

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
International Bureau



(43) International Publication Date
30 May 2002 (30.05.2002)

PCT

(10) International Publication Number
WO 02/43446 A1

(51) International Patent Classification⁷: H05B 33/14, H01L 51/20, C09K 11/06

(81) Designated States (*national*): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, TZ, UA, UG, US, UZ, VN, YU, ZA, ZW.

(21) International Application Number: PCT/GB01/05111

(22) International Filing Date:
21 November 2001 (21.11.2001)

(25) Filing Language:

English

(26) Publication Language:

English

(30) Priority Data:

0028439.8 21 November 2000 (21.11.2000) GB

(84) Designated States (*regional*): ARIPO patent (GH, GM, KE, LS, MW, MZ, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE, TR), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

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Published:

— with international search report

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For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

WO 02/43446 A1

(54) Title: ELECTROLUMINESCENT DEVICE

(57) Abstract: An electroluminescent device which emits white light which comprises sequentially an anode, a layer of a hole transporting material which emits light in the blue spectrum, a layer of an organo metallic complex and a cathode.

- 1 -

Electroluminescent Device

The present invention relates to electroluminescent materials and devices incorporating electroluminescent materials.

5

Materials which emit light when an electric current is passed through them are well known and used in a wide range of display applications. Liquid crystal devices and devices which are based on inorganic semiconductor systems are widely used, however these suffer from the disadvantages of high energy consumption, high cost 10 of manufacture, low quantum efficiency and the inability to make flat panel displays.

Organic polymers have been proposed as useful in electroluminescent devices, but it is not possible to obtain pure colours, they are expensive to make and have a relatively low efficiency.

15

Another compound which has been proposed is aluminium quinolate, but this requires dopants to be used to obtain a range of colours and has a relatively low efficiency.

20

Rare earth chelates are known which fluoresce in ultra violet radiation and A. P. Sinha (Spectroscopy of Inorganic Chemistry Vol. 2 Academic Press 1971) describes several classes of rare earth chelates with various monodentate and bidentate ligands.

25

Group III A metals and lanthanides and actinides with aromatic complexing agents have been described by G. Kallistratos (Chimica Chronika, New Series, 11, 249-266 (1982)). This reference specifically discloses the Eu(III), Tb(III), U(III) and U(IV) complexes of diphenyl-phosponamidotriphenyl-phosphoran.

30

EP 0744451A1 also discloses fluorescent chelates of transition or lanthanide or actinide metals and the known chelates which can be used are those disclosed in the

- 2 -

above references including those based on diketone and triketone moieties.

Any metal ion having an unfilled inner shell can be used as the metal and the preferred metals are selected from Sm(III), Eu(II), Eu(III), Tb(III), Dy(III), Yb(III),
5 Lu(III), Gd (III), Gd(III) U(III), Tm(III), Ce (III), Pr(III), Nd(III), Pm(III), Dy(III), Ho(III) and Er(III).

Patent application WO98/58037 describes a range of lanthanide complexes which can be used in electroluminescent devices which have improved properties and give better
10 results. Patent Applications PCT/GB98/01773, PCT/GB99/03619, PCT/GB99/04030, PCT/GB99/04024, PCT/GB99/04028, PCT/GB00/00268 describe electroluminescent complexes, structures and devices using rare earth chelates.

A typical electroluminescent device has sequentially a transparent anode such as an
15 indium tin oxide coated glass, a layer of a hole transporting material, a layer of the electroluminescent material an electron transmitting material and a metal cathode

For many applications there is a need for electroluminescent devices which emit white light or light which appears white to the eye. Hitherto in order to achieve white
20 light layers of electroluminescent materials which emit light in different colours which in combination appear white have been used. However this means a more complex device has to be made.

We have now devised an organometallic electroluminescent device which can emit
25 white light and in which the colour of the light emitted can be changed by varying the field strength applied across the device,

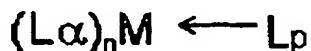
According to the invention there is provided an electroluminescent device which comprises sequentially
30 (i) a first electrode

- 3 -

- (ii) a hole transporting layer which has a component in the blue spectrum,
 - (iii) an electroluminescent layer incorporating $(L\alpha)_nM$
 - (iv) a second electrode
- 5 where M is a rare earth metal, preferably Eu, Tb, Sm or Dy and $L\alpha$ is an organic ligands and n is the valence state of M.

Preferably M is Eu, Tb, Sm or Dy.

- 10 The preferred electroluminescent compounds which can be used in the present invention are of formula



- 15 where $L\alpha$ and L_p are organic ligands. The ligands $L\alpha$ can be the same or different and there can be a plurality of ligands L_p which can be the same or different.

In this type of the ligand $L\alpha$ has a negative charge and the ligand L_p is not charged.

- 20 For example $(L_1)(L_2)(L_3)M(L_p)$ and $(L_1)(L_2)(L_3)$ are the same or different organic complexes and (L_p) is a neutral ligand. The ligands L_p can be monodentate, bidentate or polydentate and there can be one or more ligands L_p .

- Further electroluminescent compounds which can be used in the present invention are
25 of general formula $(L\alpha)_nMM_2$ where M_2 is a non rare earth metal, $L\alpha$ is as above and n is the combined valence state of M and M_2 . The complex can also comprise one or more neutral ligands L_p so the complex has the general formula $(L\alpha)_nMM_2(L_p)$, where L_p is as above. The metal M_2 can be any metal which is not a rare earth, transition metal, lanthanide or an actinide examples of metals which can be used
30 include lithium, sodium, potassium, rubidium, caesium, beryllium, magnesium,

- 4 -

calcium, strontium, barium, copper (I), copper (II), silver, gold, zinc, cadmium, boron, aluminium, gallium, indium, germanium, tin (III), tin (IV), antimony (II), antimony (IV), lead (II), lead (IV) and metals of the first, second and third groups of transition metals in different valence states e.g. manganese, iron, ruthenium, osmium, 5 cobalt, nickel, palladium(II), palladium(IV), platinum(II), platinum(IV), cadmium, chromium, titanium, vanadium, zirconium, tantalum, molybdenum, rhodium, iridium, titanium, niobium, scandium, yttrium.

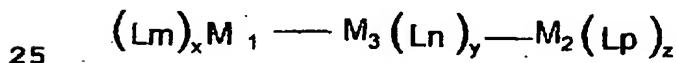
Further organometallic complexes which can be used in the present invention are
10 binuclear, trinuclear and polynuclear organometallic complexes e.g. of formula
 $(Lm)_x M_1 \leftarrow M_2 (Ln)_y$ e.g.



where L is a bridging ligand and where M_1 is M and M_2 is M or a non rare earth metal, Lm and Ln are the same or different organic ligands L as defined above, x is the valence state of M and y is the valence state of M_2 .
15

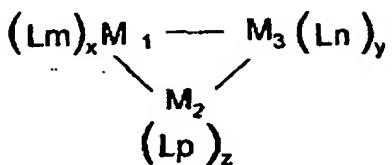
In these complexes there can be a metal to metal bond or there can be one or more bridging ligands between M_1 and M_2 and the groups Lm and Ln can be the same or
20 different.

By trinuclear is meant there are three metal atoms joined by a metal to metal bond i.e.
of formula



or

- 5 -



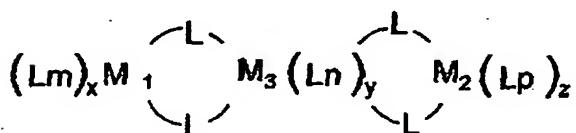
where M_1 , M_2 and M_3 are M and Lm, Ln and Lp are organic ligands Lx and y and z are all 3. Lp can be the same as Lm and Ln or different.

5

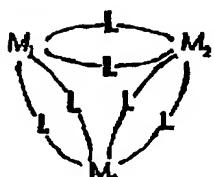
The rare earth metals and the non rare earth metals can be joined together by a metal to metal bond and/or via an intermediate bridging atom, ligand or molecular group.

For example the metals can be linked by bridging ligands e.g.

10



or

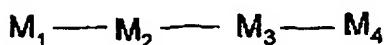


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where L is a bridging ligand

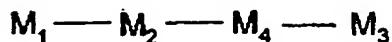
By polynuclear is meant there are more than three metal atoms joined by metal to metal bonds and/or via intermediate ligands

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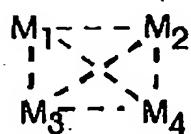


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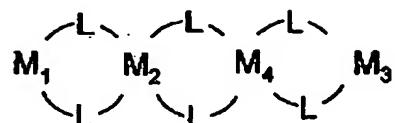
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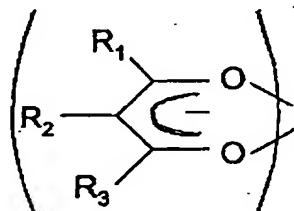
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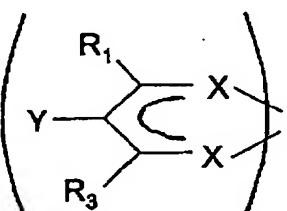
where M_1, M_2, M_3 and M_4 are M and L is a bridging ligand.

Preferably $L\alpha$ is selected from β diketones such as those of formulae

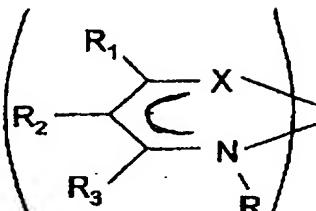
10



or



or



(I)

(II)

(III)

where R_1, R_2 and R_3 can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups; R_1, R_2 and R_3 can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene. X is Se, S or O, 15 Y can be hydrogen, substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures,

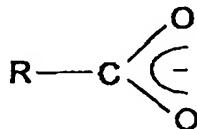
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- 7 -

fluorine, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

Examples of R₁ and/or R₂ and/or R₃ include aliphatic, aromatic and heterocyclic
5 alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.

Some of the different groups L_α may also be the same or different charged groups
10 such as carboxylate groups so that the group L₁ can be as defined above and the groups L₂, L₃... can be charged groups such as



(IV)

where R is R₁ as defined above or the groups L₁, L₂ can be as defined above and L₃...
15 etc. are other charged groups.

R₁, R₂ and R₃ can also be



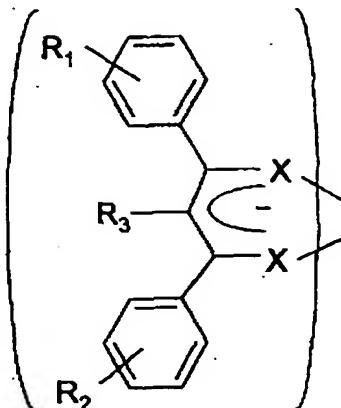
where X is O, S, Se or NH.

(V)

20 A preferred moiety R₁ is trifluoromethyl CF₃ and examples of such diketones are, benzoyltrifluoroacetone, p-chlorobenzoyltrifluoroacetone, p-bromotrifluoroacetone, p-phenyltrifluoroacetone, 1-naphthoyltrifluoroacetone, 2-naphthoyltrifluoroacetone, 2-phenathoyltrifluoroacetone, 3-phenanthroyltrifluoroacetone, 9-anthroyltrifluoroacetone, cinnamoyltrifluoroacetone, and 2-thenoyltrifluoroacetone.
25

- 8 -

The different groups $L\alpha$ may be the same or different ligands of formulae

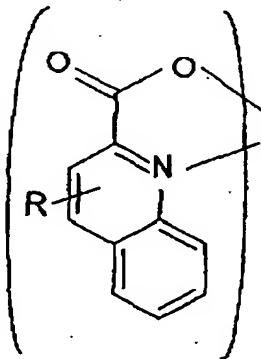


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(VI)

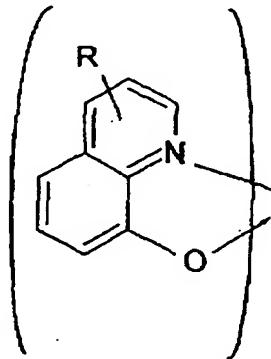
where X is O, S, or Se and R₁ R₂ and R₃ are as above

The different groups $L\alpha$ may be the same or different quinolate derivatives such as



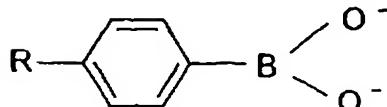
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(VII)

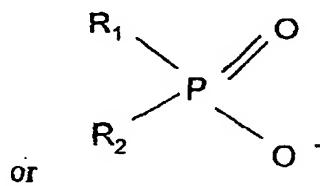


(VIII)

where R is hydrocarbyl, aliphatic, aromatic or heterocyclic carboxy, aryloxy, hydroxy or alkoxy e.g. the 8 hydroxy quinolate derivatives or



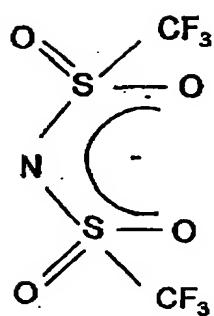
(IX)



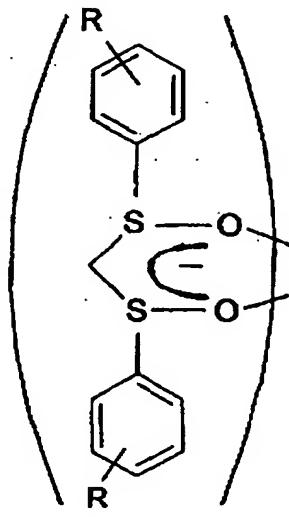
(X)

- 9 -

where R, R₁, and R₂ are as above or are H or F e.g. R₁ and R₂ are alkyl or alkoxy groups

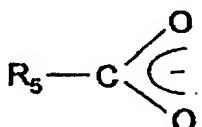


(XI)



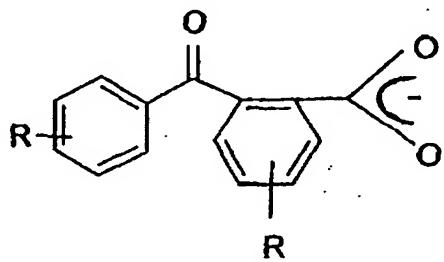
(XII)

- 5 As stated above the different groups L_a may also be the same or different carboxylate groups e.g.



(XIII)

- 10 where R₅ is a substituted or unsubstituted aromatic, polycyclic or heterocyclic ring a polypyridyl group, R₅ can also be a 2-ethyl hexyl group so L_n is 2-ethylhexanoate or R₅ can be a chair structure so that L_n is 2-acetyl cyclohexanoate or L_a can be

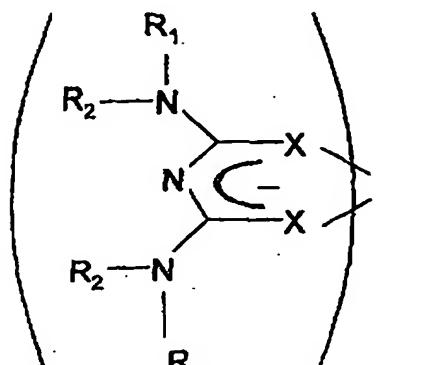


(XIV)

- 10 -

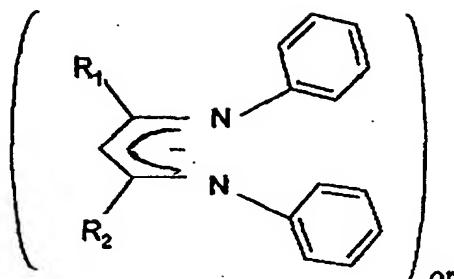
where R is as above e.g. alkyl, allenyl, amino or a fused ring such as a cyclic or polycyclic ring.

5 The different groups L_a may also be

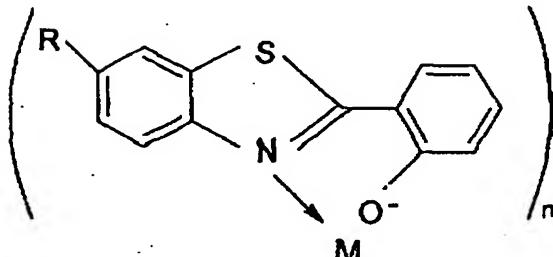


or

(XV)



(XVI)



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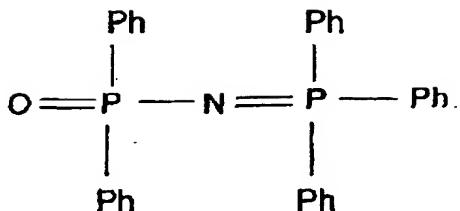
(XVII)

Where R, R₁ and R₂ are as above.

The groups L_p can be selected from

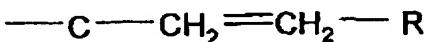
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(XVIII)

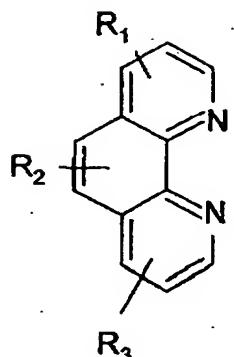
Where each Ph which can be the same or different and can be a phenyl (OPNP) or a substituted phenyl group, other substituted or unsubstituted aromatic group, a substituted or unsubstituted heterocyclic or polycyclic group, a substituted or unsubstituted fused aromatic group such as a naphthyl, anthracene, phenanthrene or pyrene group. The substituents can be for example an alkyl, aralkyl, alkoxy, aromatic, heterocyclic, polycyclic group, halogen such as fluorine, cyano, amino. Substituted amino etc. Examples are given in figs. 1 and 2 of the drawings where R, R₁, R₂, R₃ and R₄ can be the same or different and are selected from hydrogen, hydrocarbyl groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups; R, R₁, R₂, R₃ and R₄ can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene. R, R₁, R₂, R₃ and R₄ can also be unsaturated alkylene groups such as vinyl groups or groups



where R is as above.

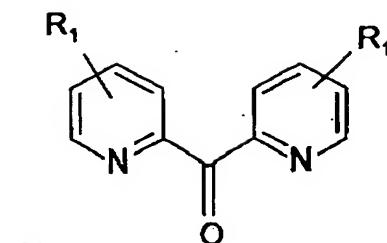
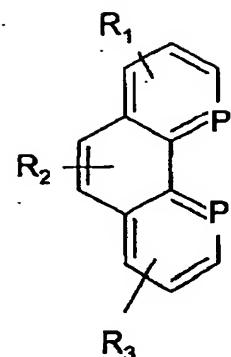
20 L_p can also be compounds of formulae

- 12 -



or

(XVIV)

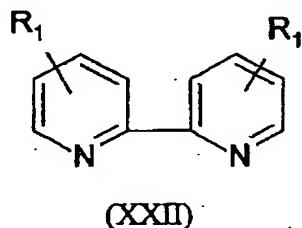


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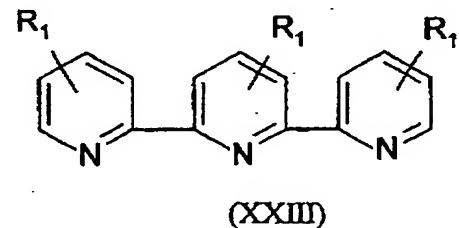
(XXI)

where R₁, R₂ and R₃ are as referred to above, for example bathophen shown in fig. 3
of the drawings in which R is as above or

5



(XXII)

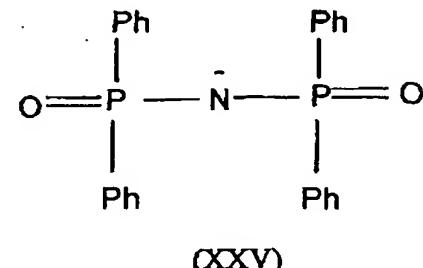
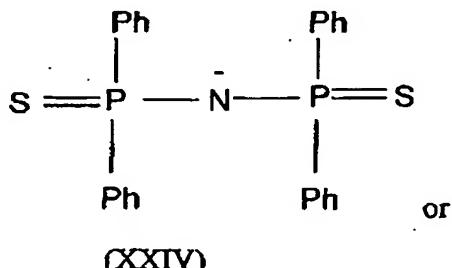


(XXIII)

where R₁, R₂ and R₃ are as referred to above.

10

L_p can also be



(XXIV)

(XXV)

15 where Ph is as above.

- 13 -

Other examples of L_p chelates are as shown in figs. 4 and fluorene and fluorene derivatives e.g. a shown in figs. 5 and compounds of formulae as shown as shown in figs. 6 to 8.

5 Specific examples of L_α and L_p are tripyridyl and TMHD, and TMHD complexes, $\alpha, \alpha', \alpha''$ tripyridyl, crown ethers, cyclans, cryptans phthalocyanans, porphoryins ethylene diamine tetramine (EDTA), DCTA, DTPA and TTHA. Where TMHD is 2,2,6,6-tetramethyl-3,5-heptanedionato and OPNP is diphenylphosphonimide triphenyl phosphorane. The formulae of the polyamines are shown in fig. 9.

10

The electroluminescent material can be deposited on the substrate directly by evaporation from a solution of the material in an organic solvent. The solvent which is used will depend on the material but chlorinated hydrocarbons such as dichloromethane, n-methyl pyrrolidone, dimethyl sulphoxide, tetra hydrofuran 15 dimethylformamide etc. are suitable in many cases.

Alternatively the material can be deposited by spin coating from solution or by vacuum deposition from the solid state e.g. by sputtering or any other conventional method can be used.

20

Preferred electroluminescent materials are Eu(DBM)₃OPNP, and tris (2,2,6,6-tetramethyl-3,5- heptanedionato) dysprosium (III) diphenyl phosphonimido triphenylphosphorane. (TTHDyOPNP).

25 By a hole transporting layer with a component in the blue spectrum is meant a hole transporting layer which emits blue light when an electric field is applied across it. Hole transporting materials which can be used are TPD, naphthylphenyldiamine (NPD) and NPB, mTDATA which have the formula shown in fig. 11 and compounds of formulae of figs. 14 of the drawings and oligomers such as oligophenylenes, 30 oligothiophenes, oligofurans.

- 14 -

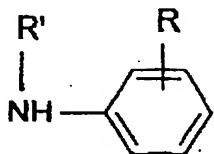
The layer can also comprise a hole transporting layer which incorporates a blue fluorescent material so that it will emit blue light and conventional blue fluorescents can be used such as tetrathiafulvene and its analogues.

5

When a blue fluorescent material is used it can be mixed with known hole transporting or hole injecting materials such as the other hole transporting materials referred to below.

- 10 There can be a layer of another hole transporting material in addition to the blue emitting hole transporting materials, these include an aromatic amine complex such as poly (vinylcarbazole), N, N'-diphenyl-N, N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD), an unsubstituted or substituted polymer of an amino substituted aromatic compound, a polyaniline, substituted polyanilines, polythiophenes, 15 substituted polythiophenes, polysilanes etc. Examples of polyanilines are polymers of

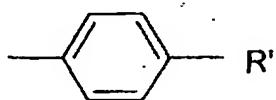
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(XXVIII)

where R is in the ortho - or meta-position and is hydrogen, C1-18 alkyl, C1-6 alkoxy, amino, chloro, bromo, hydroxy or the group

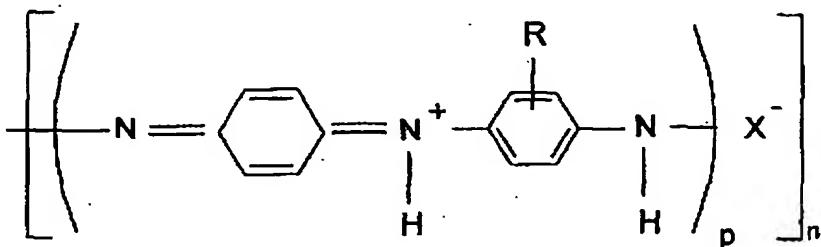
25



where R is alky or aryl and R' is hydrogen, C1-6 alkyl or aryl with at least one other monomer of formula I above.

Polyanilines which can be used in the present invention have the general formula

5



(XXIX)

where p is from 1 to 10 and n is from 1 to 20, R is as defined above and X is an anion, preferably selected from Cl, Br, SO₄, BF₄, PF₆, H₂PO₃, H₂PO₄, arylsulphonate, arenedicarboxylate, polystyrenesulphonate, polyacrylate, alkysulphonate, vinylsulphonate, vinylbenzene sulphonate, cellulosesulphonate, camphor sulphonates, cellulose sulphate or a perfluorinated polyanion.

Examples of arylsulphonates are p-toluenesulphonate, benzenesulphonate, 9,10-anthraquinone-sulphonate and anthracenesulphonate, an example of an arenedicarboxylate is phthalate and an example of arenecarboxylate is benzoate.

We have found that protonated polymers of the unsubstituted or substituted polymer of an amino substituted aromatic compound such as a polyaniline are difficult to evaporate or cannot be evaporated, however we have surprisingly found that if the unsubstituted or substituted polymer of an amino substituted aromatic compound is de-protonated it can be easily evaporated i.e. the polymer is evaporable.

Preferably evaporable de-protonated polymers of unsubstituted or substituted polymer of an amino substituted aromatic compound are used. The de-protonated unsubstituted or substituted polymer of an amino substituted aromatic compound can

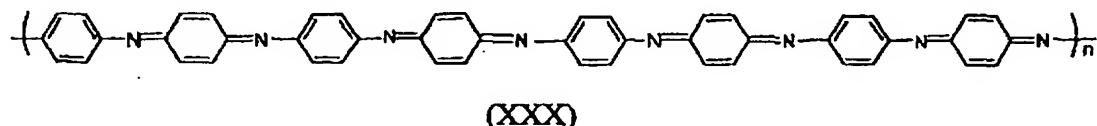
be formed by deprotonating the polymer by treatment with an alkali such as ammonium hydroxide or an alkali metal hydroxide such as sodium hydroxide or potassium hydroxide.

- 5 The degree of protonation can be controlled by forming a protonated polyaniline and de-protonating. Methods of preparing polyanilines are described in the article by A. G. MacDiarmid and A. F. Epstein, Faraday Discussions, Chem Soc.88 P319 1989.

The conductivity of the polyaniline is dependant on the degree of protonation with the
10 maximum conductivity being when the degree of protonation is between 40 and 60%
e.g. about 50% for example.

Preferably the polymer is substantially fully de-protonated

- 15 A polyaniline can be formed of octamer units i.e. p is four e.g.



The polyanilines can have conductivities of the order of 1×10^1 Siemen cm⁻¹ or higher.

20

The aromatic rings can be unsubstituted or substituted e.g. by a C1 to 20 alkyl group such as ethyl.

- 25 The polyaniline can be a copolymer of aniline and preferred copolymers are the copolymers of aniline with o-anisidine, m-sulphanilic acid or o-aminophenol, or o-toluidine with o-aminophenol, o-ethylaniline, o-phenylene diamine or with amino anthracenes.

Other polymers of an amino substituted aromatic compound which can be used include substituted or unsubstituted polyaminonaphthalenes, polyaminoanthracenes, polyaminophenanthrenes, etc. and polymers of any other condensed polyaromatic compound. Polyaminoanthracenes and methods of making them are disclosed in US 5 Patent 6,153,726. The aromatic rings can be unsubstituted or substituted e.g. by a group R as defined above.

The polyanilines can be deposited on the first electrode by conventional methods e.g. by vacuum evaporation, spin coating, chemical deposition, direct electrodeposition etc. preferably the thickness of the polyaniline layer is such that the layer is conductive and transparent and can be preferably from 20nm to 200nm. The polyanilines can be protonated or unprotonated, when they are protonated they can be dissolved in a solvent and deposited as a film, when they are unprotonated they are solids and can be deposited by vacuum evaporation i.e. by sublimation.

15

The polymers of an amino substituted aromatic compound such as polyanilines referred to above can also be used as buffer layers with other hole transporting materials.

20

The structural formulae of some other hole transmitting materials are shown in Figures 11, 12, 13 and 14 of the drawings, where R₁, R₂ and R₃ can be the same or different and are selected from hydrogen, and substituted and unsubstituted hydrocarbyl groups such as substituted and unsubstituted aliphatic groups, substituted and unsubstituted aromatic, heterocyclic and polycyclic ring structures, fluorocarbons such as trifluoryl methyl groups, halogens such as fluorine or thiophenyl groups; R₁, R₂ and R₃ can also form substituted and unsubstituted fused aromatic, heterocyclic and polycyclic ring structures and can be copolymerisable with a monomer e.g. styrene. X is Se, S or O, Y can be hydrogen, substituted or unsubstituted hydrocarbyl groups, such as substituted and unsubstituted aromatic, heterocyclic and polycyclic

ring structures, fluorine, fluorocarbons such as trifluoromethyl groups, halogens such as fluorine or thiophenyl groups or nitrile.

5 Examples of R₁ and/or R₂ and/or R₃ include aliphatic, aromatic and heterocyclic alkoxy, aryloxy and carboxy groups, substituted and substituted phenyl, fluorophenyl, biphenyl, phenanthrene, anthracene, naphthyl and fluorene groups alkyl groups such as t-butyl, heterocyclic groups such as carbazole.

Other hole transporting materials which can be used are conjugated polymers.

10 US Patent 5807627 discloses an electroluminescence device in which there are conjugated polymers in the electroluminescent layer. The conjugated polymers referred to are defined as polymers for which the main chain is either fully conjugated possessing extended pi molecular orbitals along the length of the chain or else is substantially conjugated, but with interruptions to conjugation, either random or regular along the main chain. They can be homopolymers or copolymers.

15 The conjugated polymer used can be any of the conjugated polymers disclosed or referred to in US 5807627, PCT/WO90/13148 and PCT/WO92/03490.

20 The conjugated polymers disclosed are poly (p-phenylenevinylene)-PPV and copolymers including PPV. Other preferred polymers are poly(2,5 dialkoxyphenylene vinylene) such as poly (2-methoxy-5-(2-methoxypentyloxy-1,4-phenylene vinylene), poly(2-methoxypentyloxy)-1,4-phenylenevinylene), poly(2-methoxy-5-(2-25 dodecyloxy-1,4-phenylenevinylene) and other poly(2,5 dialkoxyphenylenevinylene)s with at least one of the alkoxy groups being a long chain solubilising alkoxy group, poly fluorenes and oligofluorenes, polyphenylenes and oligophenylenes, polyanthracenes and oligo anthracenes, polythiophenes and oligothiophenes.

In PPV the phenylene ring may optionally carry one or more substituents e.g. each independently selected from alkyl, preferably methyl, alkoxy, preferably methoxy or ethoxy.

- 5 Any poly(arylenevinylene) including substituted derivatives thereof can be used and the phenylene ring in poly(p-phenylenevinylene) may be replaced by a fused ring system such as anthracene or naphthlyene ring and the number of vinylene groups in each polyphenylenevinylene moiety can be increased e.g. up to 7 or higher.
- 10 The conjugated polymers which emit light in the blue spectrum can be used as the blue hole transporting layer.

The conjugated polymers can be made by the methods disclosed in US 5807627, PCT/WO90/13148 and PCT/WO92/03490.

- 15 The hole transporting material can optionally be mixed with the electroluminescent material in a ratio of 5 - 95% of the electroluminescent material to 95 to 5% of the hole transporting compound.
- 20 The first electrode is preferably a transparent substrate which is a conductive glass or plastic material which acts as the cathode, preferred substrates are conductive glasses such as indium tin oxide coated glass, but any glass which is conductive or has a conductive layer can be used. Conductive polymers and conductive polymer coated glass or plastics materials can also be used as the substrate. The first electrode can comprise a transparent metal such as gold, silver a platinum group metal etc.

In general the thickness of the layers is from 5nm to 500nm.

- 30 The second electrode functions as the cathode and can be any low work function metal e.g. aluminium, calcium, lithium, silver/magnesium alloys etc., aluminium is a

preferred metal.

Optionally there is a layer of an electron transporting material between the second electrode, which electrode functions as the cathode, and the electroluminescent material layer, the electron transmitting material is a material which will transport electrons when an electric current is passed through electron transmitting materials include a metal complex such as a metal quinolate e.g. an aluminium quinolate, lithium quinolate a cyano anthracene such as 9,10 dicyano anthracene, a polystyrene sulphonate and compounds of formulae shown in Fig. 10. Instead of being a separate layer the electron transmitting material can be mixed with the electroluminescent material to form one layer e.g. in a proportion of 5 to 95% of the electron transmitting material to 95 to 5% of the light emitting metal compound.

The electroluminescent layer can comprise a mixture of the light emitting metal compound with the hole transporting material and electron transmitting material

The electroluminescent material can be deposited on the substrate directly by vacuum evaporation or evaporation from a solution in an organic solvent. The solvent which is used will depend on the material but chlorinated hydrocarbons such as dichloromethane and n-methyl pyrrolidone; dimethyl sulphoxide; tetra hydrofuran; dimethylformamide etc. are suitable in many cases.

Alternatively electroluminescent material can be deposited by spin coating from solution, or by vacuum deposition from the solid state e.g. by sputtering, or any other conventional method can be used.

Preferably the first electrode is a transparent substrate such as a conductive glass or plastic material which acts as the anode, preferred substrates are conductive glasses such as indium tin oxide coated glass, but any glass which is conductive or has a transparent conductive layer such as a metal or conductive polymer can be used.

Conductive polymers and conductive polymer coated glass or plastics materials can also be used as the substrate.

Either or both electrodes can be formed of silicon and the electroluminescent material 5 and intervening layers of a hole transporting and electron transporting materials can be formed as pixels on the silicon substrate.

Preferably, the substrate is of crystalline silicon and the surface of the substrate may be polished or smoothed to produce a flat surface prior to the deposition of electrode, 10 or electroluminescent compound. Alternatively a non-planarised silicon substrate can be coated with a layer of conducting polymer to provide a smooth, flat surface prior to deposition of further materials.

In one embodiment, each pixel comprises a metal electrode in contact with the 15 substrate. Depending on the relative work functions of the metal and transparent electrodes, either may serve as the anode with the other constituting the cathode.

When the silicon substrate is the cathode an indium tin oxide coated glass can act as the anode and light is emitted through the anode. When the silicon substrate acts as 20 the anode the cathode can be formed of a transparent electrode which has a suitable work function, for example by a indium zinc oxide coated glass in which the indium zinc oxide has a low work function. The anode can have a transparent coating of a metal formed on it to give a suitable work function. These devices are sometimes referred to as top emitting devices or back emitting devices.

25 The metal electrode may consist of a plurality of metal layers, for example a higher work function metal such as aluminium deposited on the substrate and a lower work function metal such as calcium deposited on the higher work function metal. In another example, a further layer of conducting polymer lies on top of a stable metal

such as aluminium.

Preferably, the electrode also acts as a mirror behind each pixel and is either deposited on, or sunk into, the planarised surface of the substrate. However, there
5 may alternatively be a light absorbing black layer adjacent to the substrate.

In still another embodiment, selective regions of a bottom conducting polymer layer are made non-conducting by exposure to a suitable aqueous solution allowing formation of arrays of conducting pixel pads which serve as the bottom contacts of
10 the pixel electrodes.

As described in WO00/60669 the brightness of light emitted from each pixel is preferably controllable in an analogue manner by adjusting the voltage or current applied by the matrix circuitry or by inputting a digital signal which is converted to an
15 analogue signal in each pixel circuit. The substrate preferably also provides data drivers, data converters and scan drivers for processing information to address the array of pixels so as to create images. When an electroluminescent material is used which emits light of a different colour depending on the applied voltage the colour of each pixel can be controlled by the matrix circuitry.

20 In one embodiment, each pixel is controlled by a switch comprising a voltage controlled element and a variable resistance element, both of which are conveniently formed by metal-oxide-semiconductor field effect transistors (MOSFETs) or by an active matrix transistor.

25 By changing the voltage the colour emitted can be changed and, as it possible to have very rapid controlled changes in voltage, this enables there to be a device which can have a very rapid change in colour in the light emitted. By a suitable construction, in which the voltage can be controlled at different locations, it is possible to have a planar device in which different colour light can be emitted at different locations and
30

the colour of the emitted light can be varied rapidly. This enables a wide range of controlled display devices emitting different colours to be constructed.

Example 1 Device Fabrication

5

An ITO coated glass piece ($1 \times 1\text{cm}^2$) had a portion etched out with concentrated hydrochloric acid to remove the ITO and was cleaned and dried. The device was fabricated by sequentially forming on the ITO layers comprising ITO/TPD(6mg)/Eu(DBM)₃OPNP(5mg)/Al by vacuum evaporation. Where TPD as
10 defined herein.

The organic coating on the portion which had been etched with, the concentrated hydrochloric acid was wiped with a cotton bud.

15 The coated electrodes were stored in a vacuum desiccator over a molecular sieve and phosphorous pentoxide until they were loaded into a vacuum coater (Edwards, 10^{-6} torr) and aluminium top contacts made. The active area of the LED's was 0.08 cm^2 by 0.1 cm^2 the devices were then kept in a vacuum desiccator until the electroluminescence studies were performed.

20 The ITO electrode was always connected to the positive terminal. The current vs. voltage studies were carried out on a computer controlled Keithly 2400 source meter.

25 Electroluminescence spectra were recorded by means of a computer controlled charge coupled device on PR650 system made by Photoresearch Inc.

30 The voltage was increased and the colour emitted noted, the colours are measured in accordance with the co-ordinates in the colour chart CIE 1931. The results are shown in the graph of figs. 14 and 15 and the colour coordinates and field strength shown in Table 1.

~ 24 ~

Table 1
ITO/TPD(6mg)/Eu(DBM)₃OPNP(5mg)/Al

Colour CIE x,y		
5	1. 0.55, 0.31	0.03 cd/m ² @ 11.5V & 0.99mA
	2. 0.60, 0.31	0.09 cd/m ² @ 14.0V & 1.39mA
	3. 0.65, 0.33	0.20 cd/m ² @ 16.5V & 3.48mA
	4. 0.65, 0.33	0.46 cd/m ² @ 18.0V & 3.356mA
	5. 0.66, 0.34	1.14 cd/m ² @ 21.0V & 3.20mA
10	6. 0.66, 0.34	2.28 cd/m ² @ 22.0V & 3.33mA
	7. 0.64, 0.34	3.44 cd/m ² @ 25.0V & 3.75mA
	8. 0.62, 0.34	5.81 cd/m ² @ 28.0V & 4.51mA
	9. 0.42, 0.44	5.50 cd/m ² @ 29.0V & 4.77mA

15 Example 2

Example 1 was repeated using ITO/DFDA/(0.9mg)/TPD(6mg)/Eu(DBM)₃OPNP(6mg)/Al. DFDA is diformyl diamino anthracene and TPD is as defined herein and the results illustrated in figs.
20 16 and 17 with the colour coordinates shown in Table 2

- 25 -

Table 2

Colour CIE x,y		
	1. 0.40, 0.33	0.04 cd/m ² @ 17.5V & 0.6@A
5	2. 0.61, 0.33	0.05 cd/m ² @ 20.0V & 1.5@A
	3. 0.63, 0.33	0.15 cd/m ² @ 22.5V & 5.3@A
	4. 0.66, 0.33	0.43 cd/m ² @ 26.0V & 15.7@A
	5. 0.66, 0.34	1.08 cd/m ² @ 29.0V & 48@A
	6. 0.66, 0.33	1.13 cd/m ² @ 31.0V & 47@A
10	7. 0.66, 0.34	3.47 cd/m ² @ 34.0V & 189@A
	8. 0.65, 0.34	6.45 cd/m ² @ 36.0V & 357@A
	9. 0.64, 0.34	12.1 cd/m ² @ 40.0V & 6651@A

Example 3 Preparation of Tris (2,2,6,6- tetramethyl-3,5- heptanedionato) dysprosium (III) diphenyl phosphonimido triphenylphosphorane. (TTHDyOPNP)

Tris (2,2,6,6- tetramethyl-3,5- heptanedionato) dysprosium (III) (6.1g, 19.5 mmole) and diphenyl phosphonimido triphenylphosphorane. (4.6g, 9.5 mmole) were refluxed in trimethylpentane (60ml) for 30 minutes . The reaction mixture was the allowed to cool to room temperature. A white crystalline material formed on standing. This was recrystallised from diethyl ether to give tris (2,2,6,6- tetramethyl-3,5- heptanedionato) dysprosium (III) diphenyl phosphonimido triphenylphosphorane yield : 8 gm., mpt.154°C)

25

Example 4 Device Fabrication

An ITO coated glass piece (1 x 1cm²) had a portion etched out with concentrated hydrochloric acid to remove the ITO and was cleaned and dried. The device was

fabricated by sequentially forming on the ITO, by vacuum evaporation, layers comprising:-

ITO /DFDAA (5nm) / mMTDATA(30 nm)/ TTHDyOPNP (55nm) /LiF (0.7nm)/Al(105nm)

5

Where ITO is indium titanium oxide coated glass mMTDATA and DFDAA are as defined herein.

The organic coating on the portion which had been etched with the concentrated
10 hydrochloric acid was wiped with a cotton bud. The coated electrodes were stored in a vacuum desiccator over a molecular sieve and phosphorous pentoxide until they were loaded into a vacuum coater (Edwards, 10^{-6} torr) and aluminium top contacts made. The active area of the LED's was 0.08 cm by 0.1 cm² the devices were then kept in a vacuum desiccator until the electroluminescence studies were performed.

15

The ITO electrode was always connected to the positive terminal. The current vs. voltage studies were carried out on a computer controlled Keithly 2400 source meter.

20 An electric current was applied across the device and the spectrum shown in fig. 18 is a plot of the current versus wavelength is shown in fig. 19.

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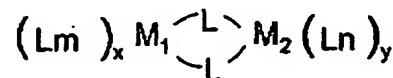
Claims

1. An electroluminescent device which comprises sequentially
 - (i) a first electrode
 - 5 (ii) a hole transporting layer which has a component in the blue spectrum,
 - (iii) an electroluminescent layer incorporating $M(L\alpha)_n$ a second electrode where M , is a rare earth metal and $(L\alpha)$ is an organic ligand.
- 10 2. An electroluminescent device as claimed in claim 1 in which the electroluminescent layer incorporates a compound of formula
$$(L\alpha)_nM \leftarrow L_p$$
where $L\alpha$ and L_p are organic ligands, the ligands $L\alpha$ can be the same or different and
15 there can be a plurality of ligands L_p which can be the same or different.
- 20 3. An electroluminescent device as claimed in claim 2 in which the electroluminescent compound is a complex of formula $(L_1)(L_2)(L_3)M(L_p)$ and $(L_1)(L_2)(L_3)$ are the same or different organic complexes $L\alpha$ and (L_p) is a neutral ligand.
- 25 4. An electroluminescent device as claimed in claim 2 in which the electroluminescent compound is a complex of formula $(L\alpha)_nMM_2$ where M_2 is a non rare earth metal, $L\alpha$ is as above and n is the combined valence state of M and M_2 .
5. An electroluminescent device as claimed in claim 2 in which the electroluminescent compound is a complex of formula $(L\alpha)_nMM_2(L_p)$, where L_p is as above and the metal M_2 is any metal which is not a rare earth, transition metal, lanthanide or an actinide.

- 28 -

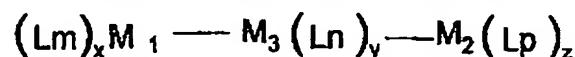
6. An electroluminescent device as claimed in claim 2 in which the electroluminescent compound is a metal complex of formula

$(Lm)_x M_1 \leftarrow M_2 (Ln)_y$ or

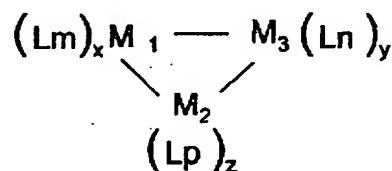


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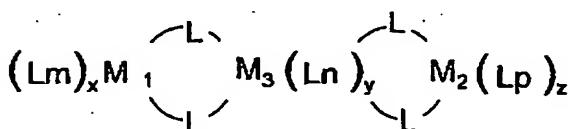
where L is a bridging ligand and where M_1 is M and M_2 is M or a non rare earth metal, Lm and Ln are the same or different organic ligands La as defined above, x is the valence state of M and y is the valence state of M_2 or of formula



10 or

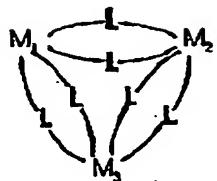


15 where M_1 , M_2 and M_3 are M and Lm, Ln and Lp are organic ligands La and x, y and z are all 3, Lp can be the same as Lm and Ln or different and the rare earth metals and the non rare earth metals are joined together by a metal to metal bond and/or via an intermediate bridging atom, ligand or of formula

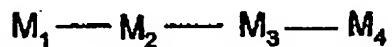


20 or

- 29 -



where L is a bridging ligand or of formula

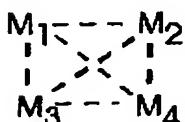


or

5

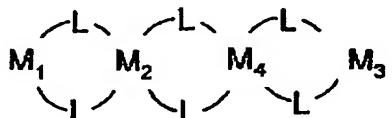


or



10

or



where M_1 , M_2 , M_3 and M_4 are M and L is a bridging ligand.

7. An electroluminescent device as claimed in any one of claims 1 to 6 in which L_a

15 has a formula of (I) to (XVII) herein.

8. An electroluminescent device as claimed in any one of claims 1 to 7 in which L_p
has a formula of (XVIII) to (XXV) or as in figs. 1 to 8 herein.

20 9. An electroluminescent device as claimed in any one of claims 1 to 8 in which the
hole transporting material is mTDATA, TPD, NPD, NPV, as herein defined or has a
formula as set out in figs 11, 12 or 13

- 30 -

10. An electroluminescent device as claimed in any one of claims 1 to 9 in which M is Eu, Tb, Sm or Dy
11. An electroluminescent device as claimed in any one of claims 1 to 10 in which the
5 hole transporting layer incorporates a blue fluorescent.
12. An electroluminescent device as claimed in any one of claims 1 to 11 in which the thickness of the hole transporting layer is greater than 5nm.
- 10 13. An electroluminescent device as claimed in claim 12 in which the thickness of the hole transporting layer is from 10 to 50nm.
14. An electroluminescent device as claimed in any one of claims 1 to 13 in which the thickness of the electroluminescent layer is greater than 5nm
- 15 15. An electroluminescent device as claimed in any one of claims 1 to 14 in which the thickness of the electroluminescent layer is from 10 to 40nm.
16. An electroluminescent device as claimed in any one of the preceding claims in
20 which the second electrode is aluminium, calcium, lithium, or a silver/magnesium alloy.
17. An electroluminescent device as claimed in any one of the preceding claims in
which there is another organic hole transporting layer in contact with the layer of light
25 emitting material.
18. An electroluminescent device as claimed in any one of claims 1 to 17 in which the hole transporting material is a film of a polymer selected from poly(vinylcarbazole),
N,N'-diphenyl-N,N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD),
30 unsubstituted or substituted polymer of an amino substituted aromatic compounds,

- 31 -

polyaniline, substituted polyanilines, polythiophenes, substituted polythiophenes, polysilanes and substituted polysilanes, de-protonated unsubstituted or substituted polymers of an amino substituted aromatic compounds, de-protonated polyanilines

- 5 19. An electroluminescent device as claimed in claim 17 in which the other hole transporting material is a film of a compound selected from poly(vinylcarbazole), N,N'-diphenyl-N,N'-bis (3-methylphenyl) -1,1' -biphenyl -4,4'-diamine (TPD), unsubstituted or substituted polymer of an amino substituted aromatic compounds, polyaniline, substituted polyanilines, polythiophenes, substituted polythiophenes, polysilanes and substituted polysilanes, de-protonated unsubstituted or substituted polymers of an amino substituted aromatic compounds, de-protonated polyanilines of formula (XXIX) or (XXX) herein or as in Figure 11, 12, 13, or 14 of the drawings or is a conjugated polymer.
- 10 20. An electroluminescent device as claimed in any one of claims 1 to 19 in which a hole transporting material and the light emitting metal compound are mixed to form one layer.
- 15 21 A device as claimed in any one of claims 1 to 20 in which there is a layer of an electron transmitting material between the second electrode and the layer of the electroluminescent complex
- 20 22. An electroluminescent device as claimed in claim 21 in which the electron transmitting material is a metal quinolate or as in fig. 10 of the drawings.
- 25 23. An electroluminescent device as claimed in claim 22 in which the electron transmitting material is lithium quinolate.

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24. An electroluminescent device as claimed in any one of claims 21 to 23 in which an electron transmitting material and the light emitting metal compound are mixed to form one layer.
- 5 25. An electroluminescent device as claimed in any one of the preceding claims in which the second electrode is selected from aluminium, calcium, lithium, and silver/magnesium alloys.

1/17

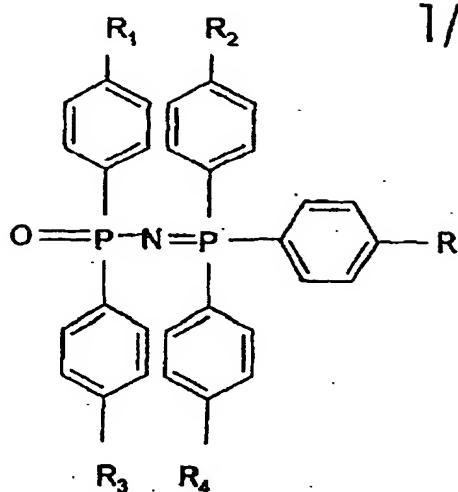


Fig. 1

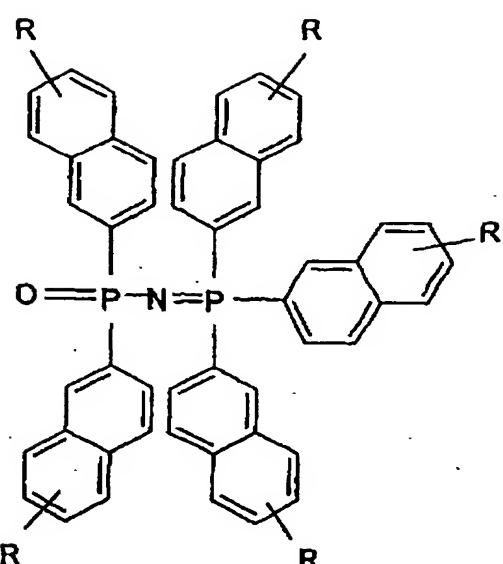


Fig. 2a

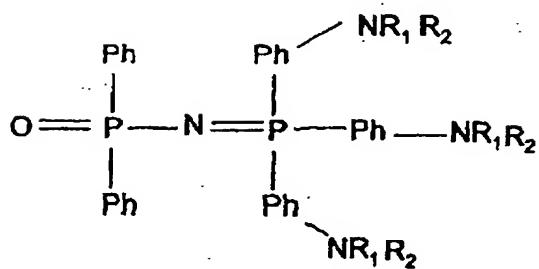


Fig. 2b

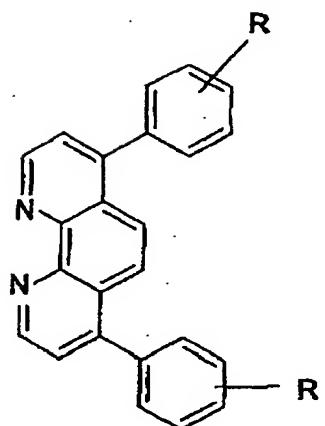


Fig. 3

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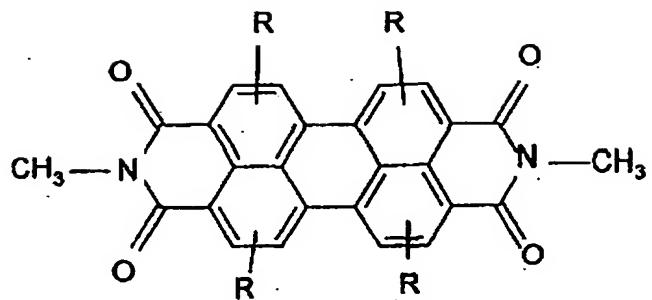


Fig. 4a

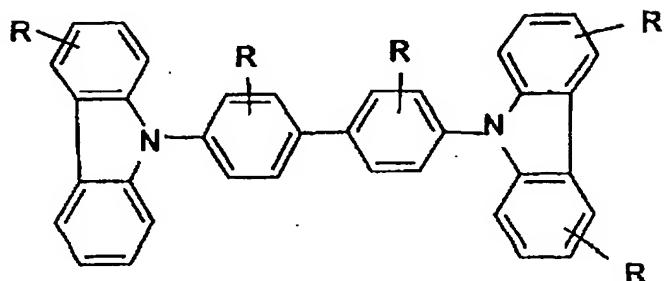


Fig. 4b

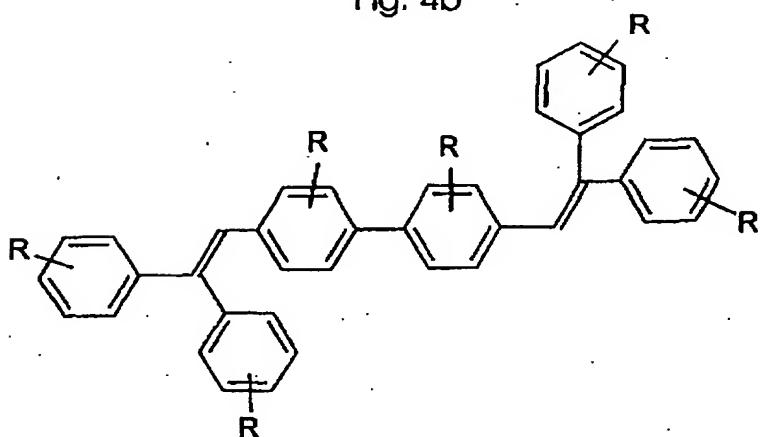


Fig. 4c

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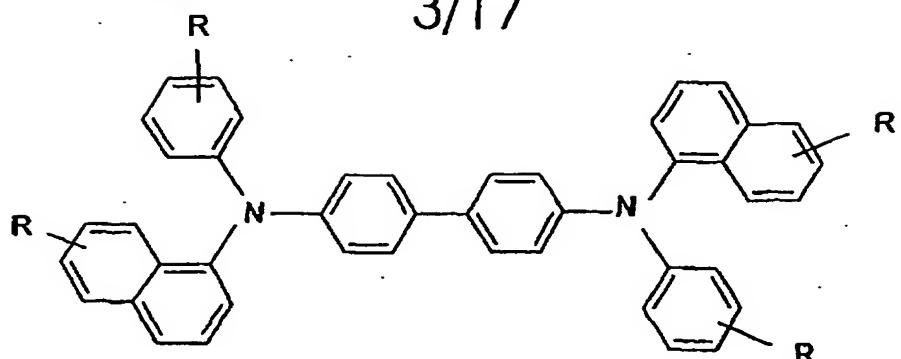


Fig. 4d

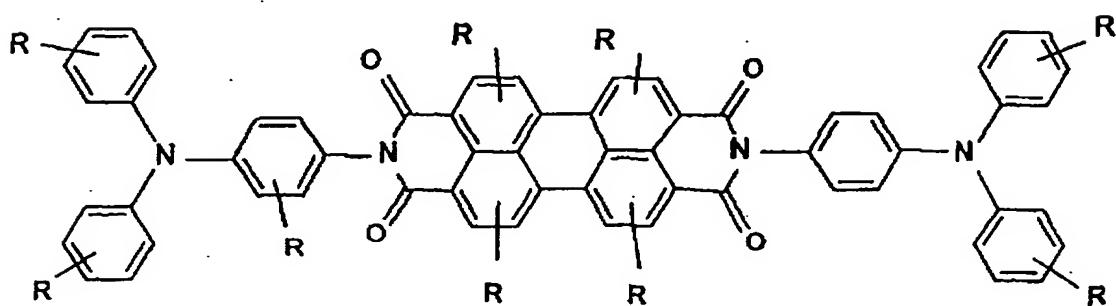


Fig. 4e

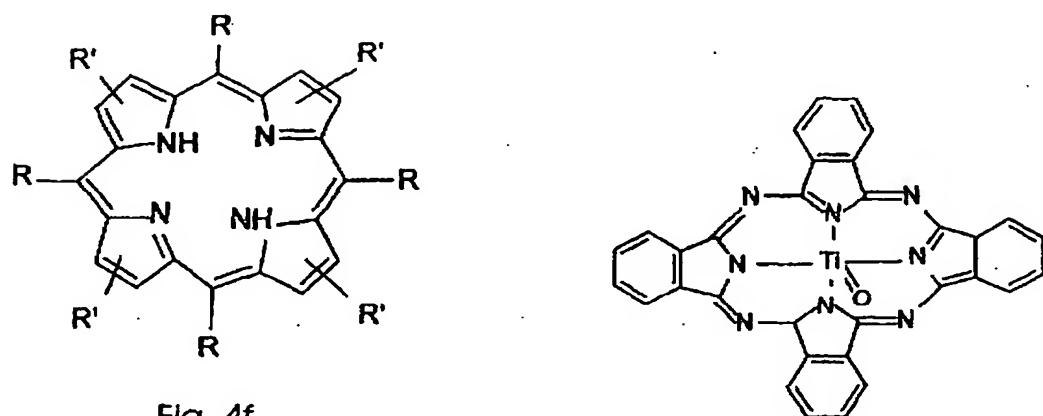


Fig. 4f

Fig. 4g

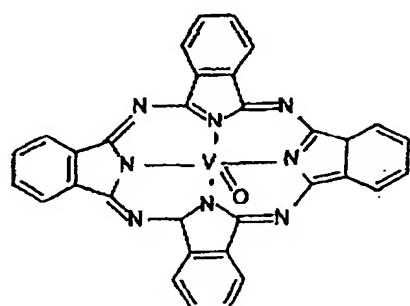


Fig. 4h

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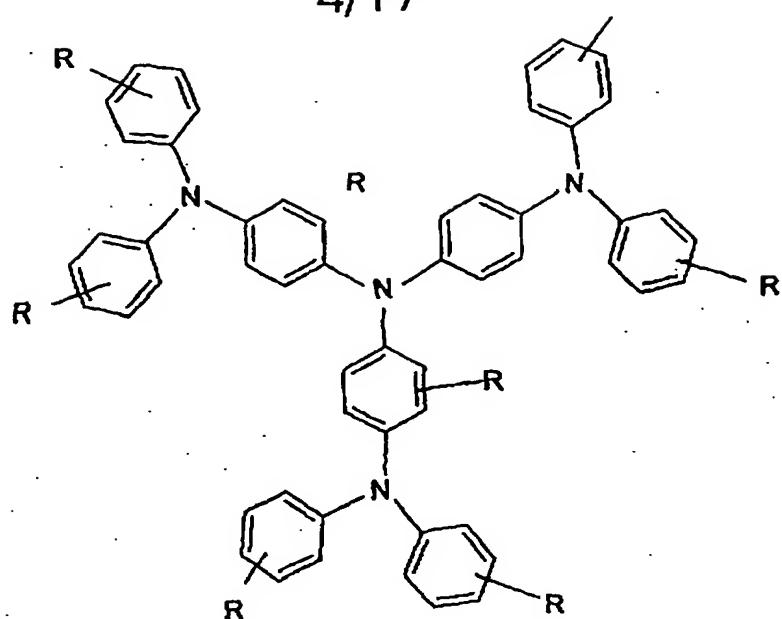


Fig. 4i

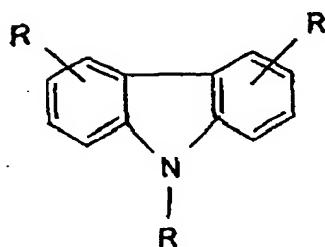


Fig. 4j

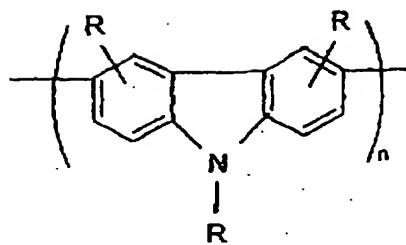


Fig. 4k

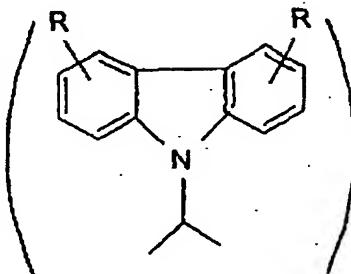


Fig. 4l

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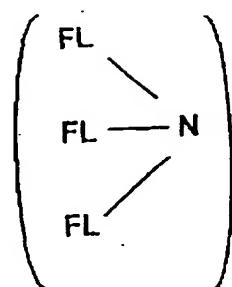


Fig. 5a

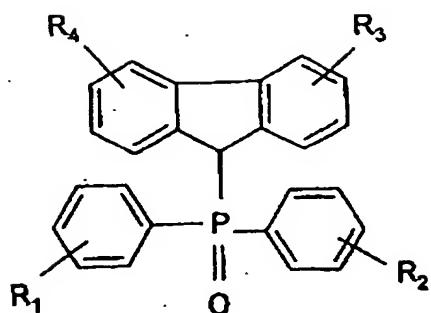


Fig. 5b

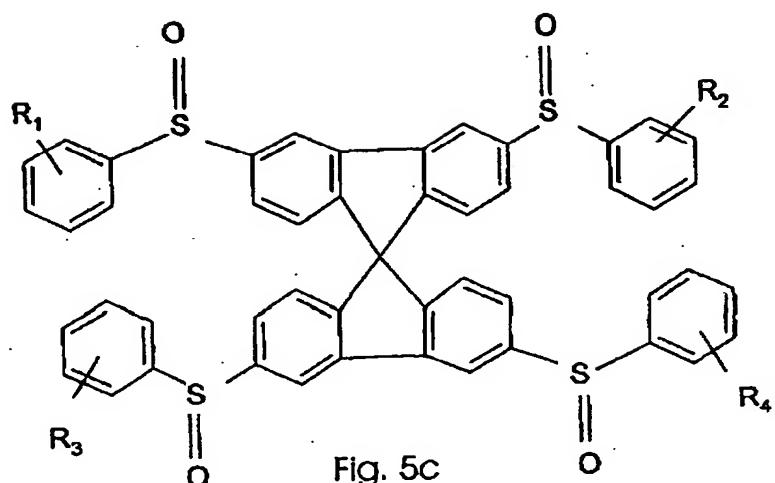


Fig. 5c

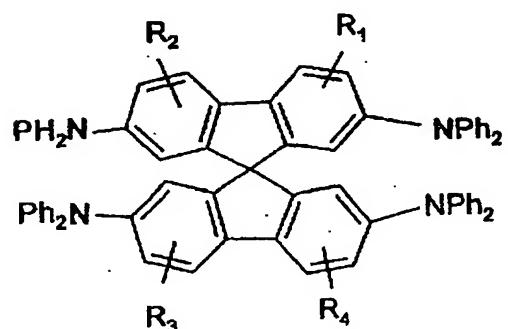


Fig. 5d

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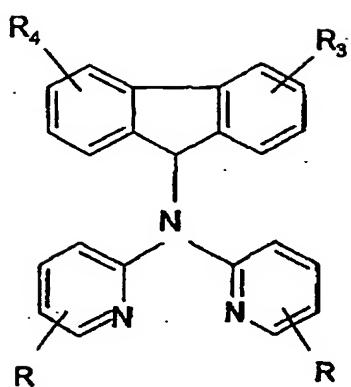


Fig. 5f

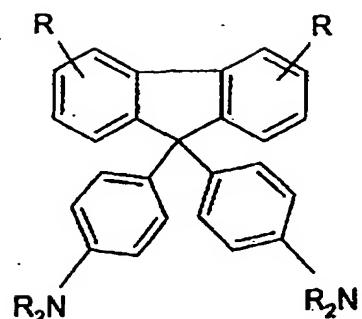


Fig. 5g

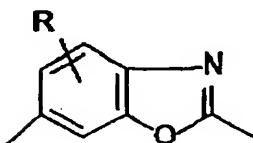


Fig. 6a

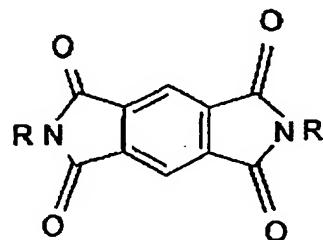


Fig 6b

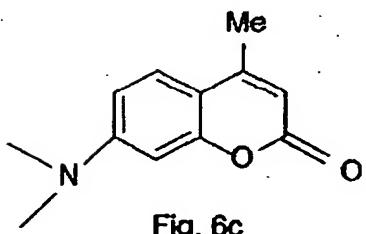


Fig. 6c

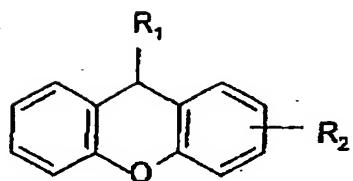


Fig. 6d

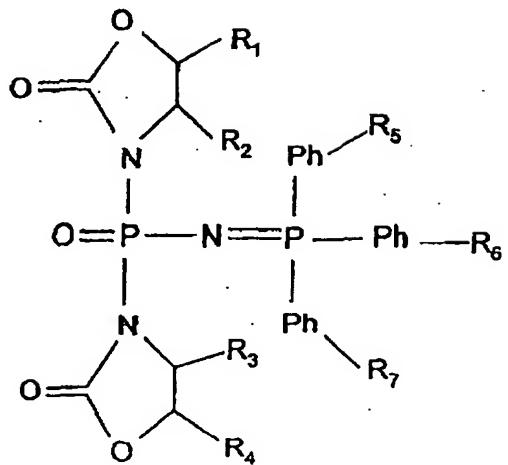
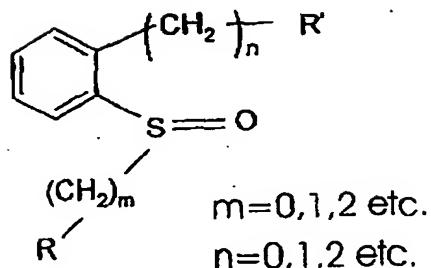
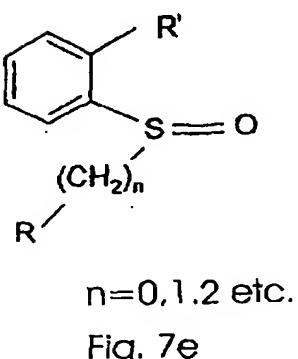
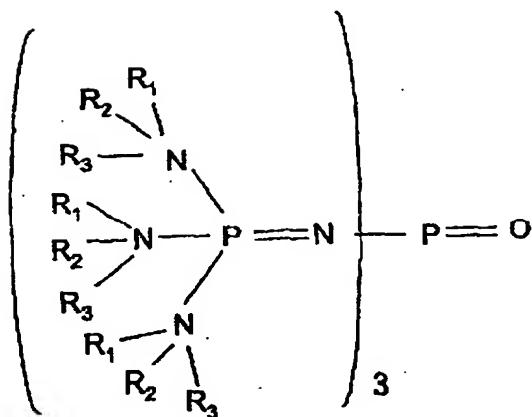
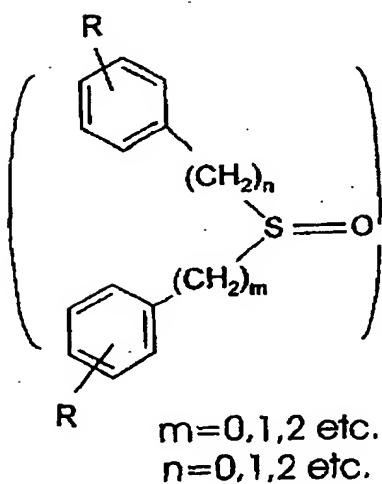
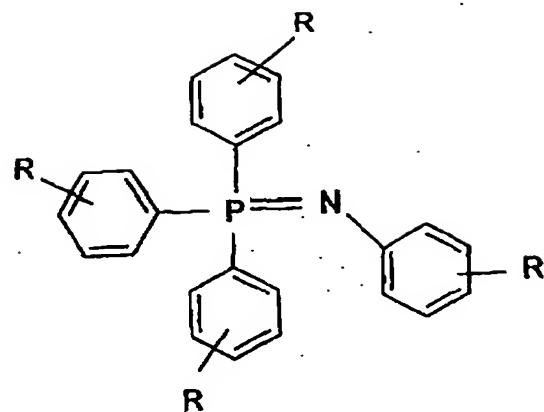
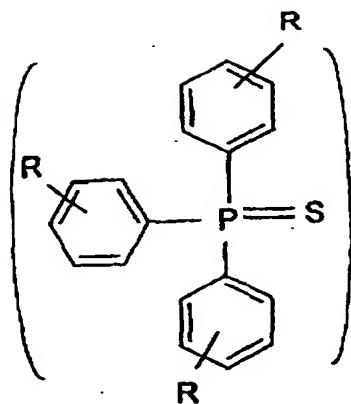


Fig. 6e

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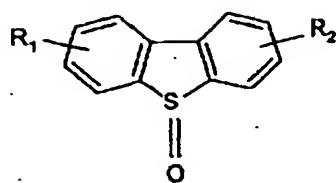


Fig. 8a

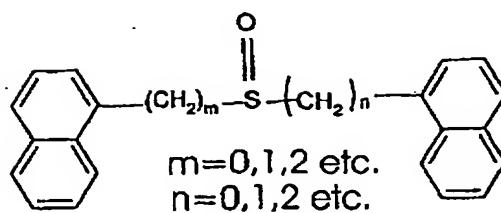


Fig. 8b

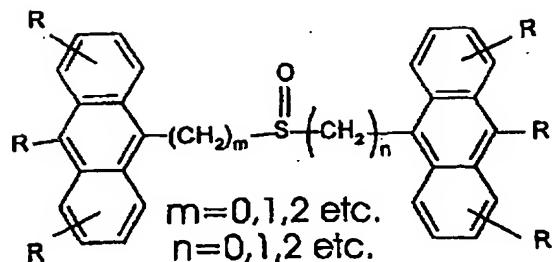


Fig. 8c

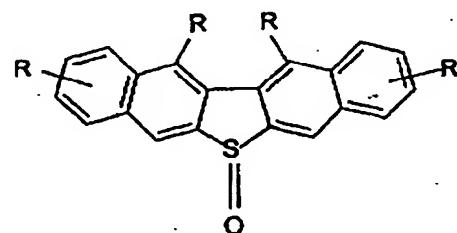


Fig. 8d

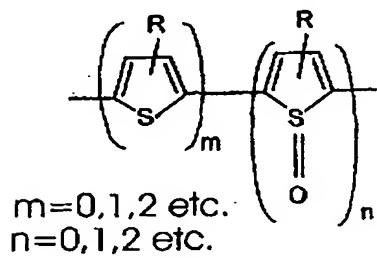


Fig. 8e

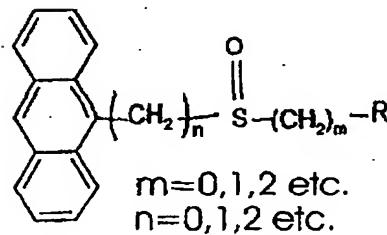


Fig. 8f

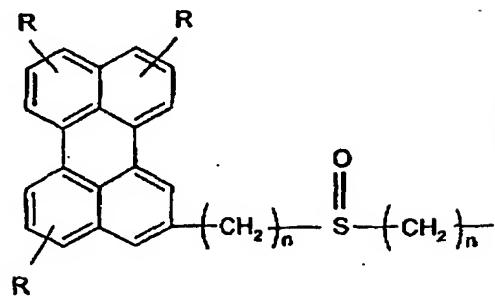


Fig. 8g

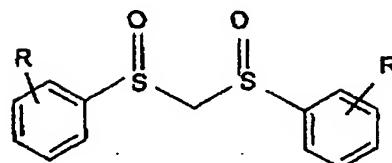
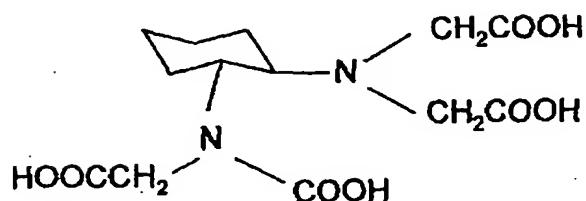
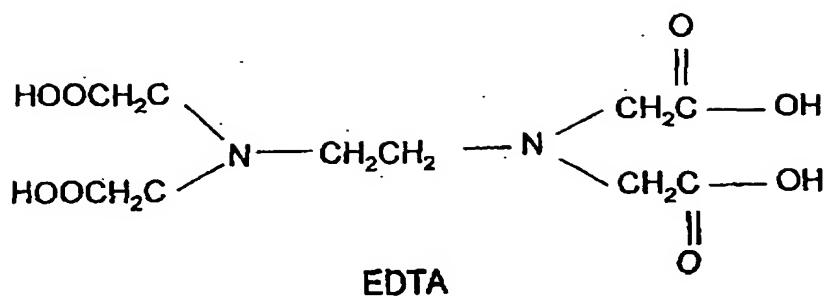
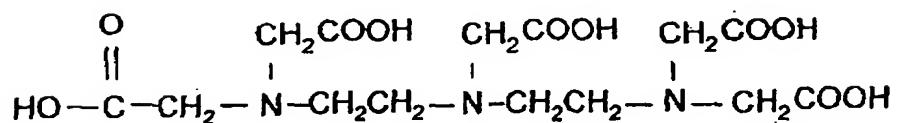


Fig. 8h

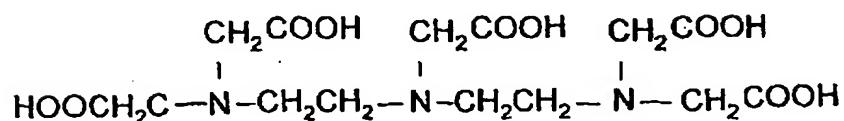
9/17



DCTA



DTPA



TTHA

Fig. 9

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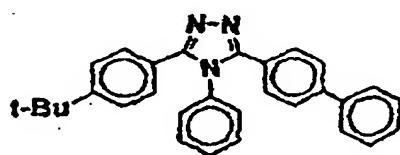
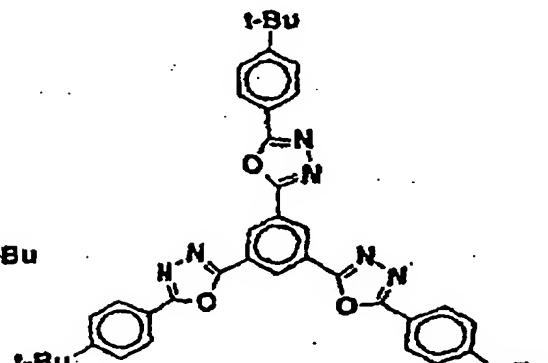
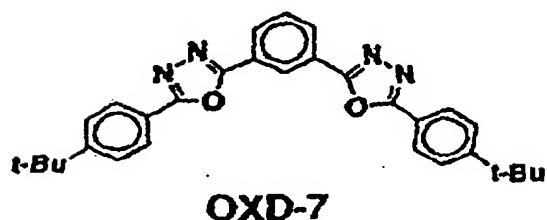
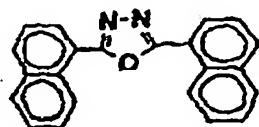
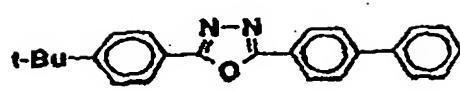
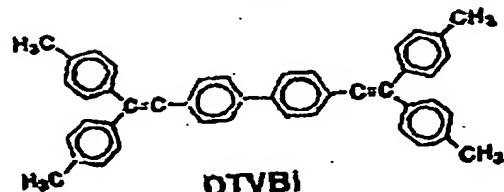
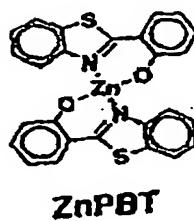
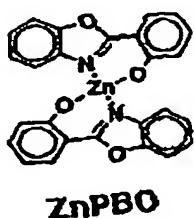
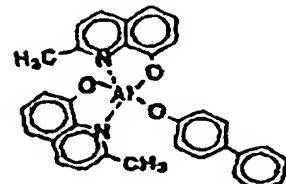
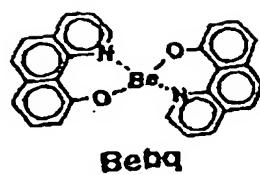
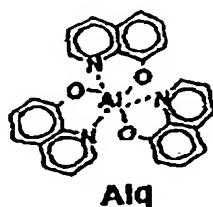
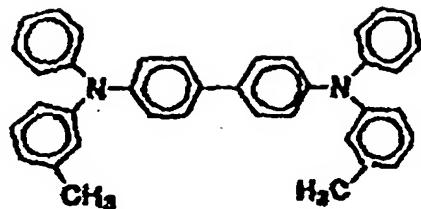


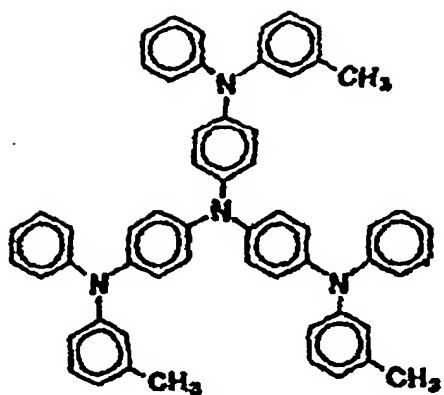
Fig. 10

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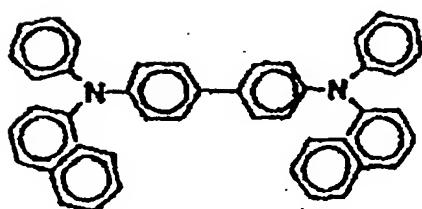
11/17



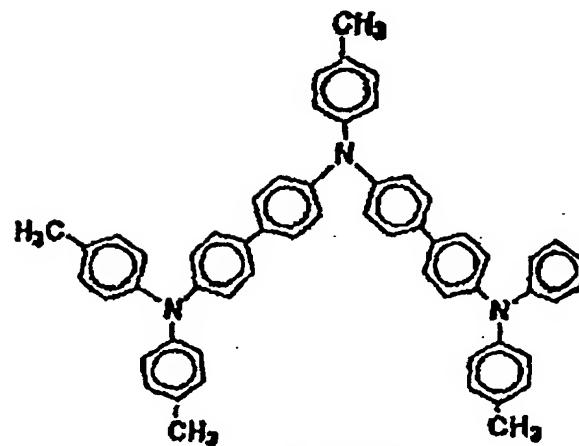
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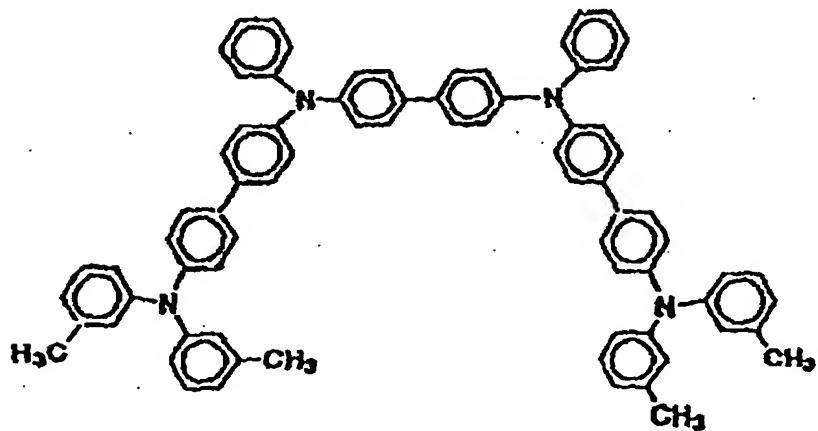
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α-NPD



HTM-1



TPTE

Fig. 11

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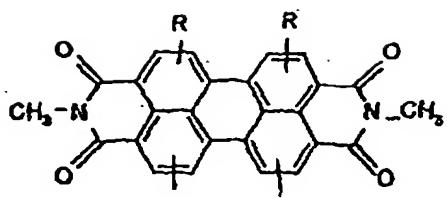


Fig. 12a

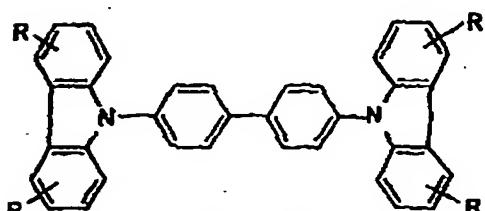


Fig. 12b

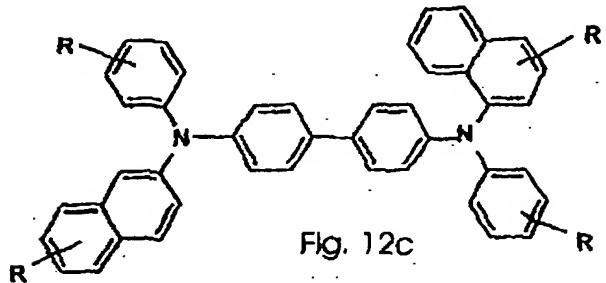


Fig. 12c

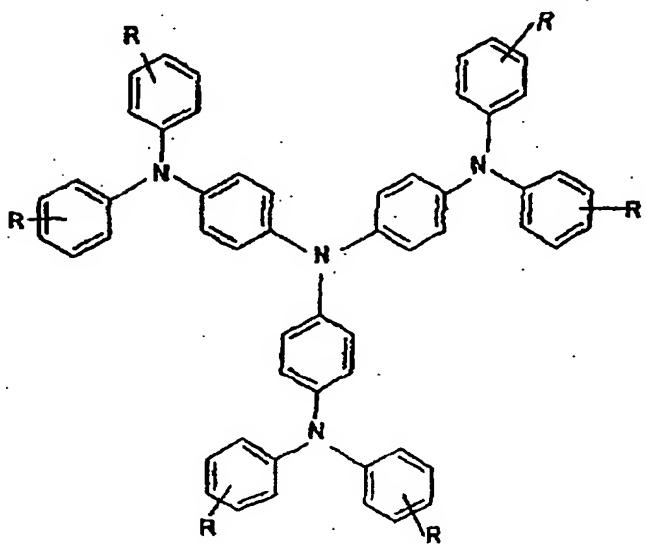


Fig. 12d

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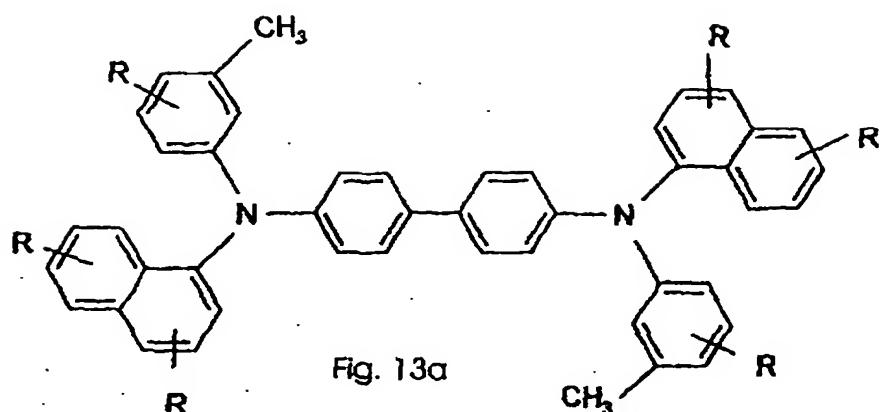


Fig. 13a

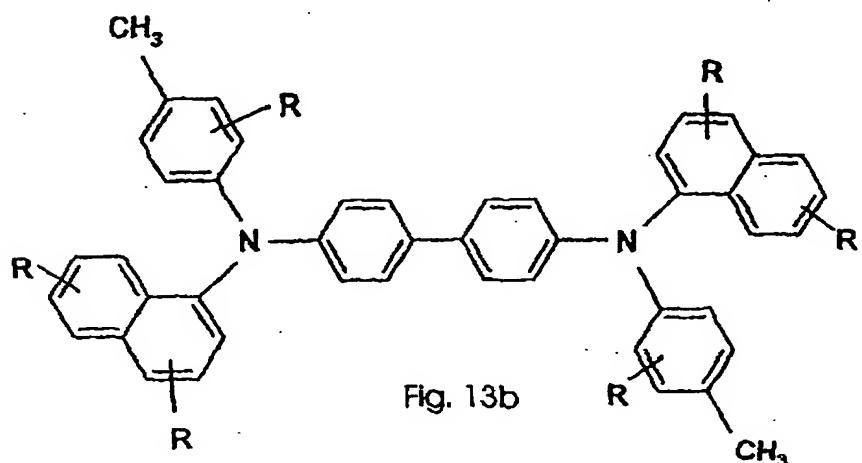


Fig. 13b

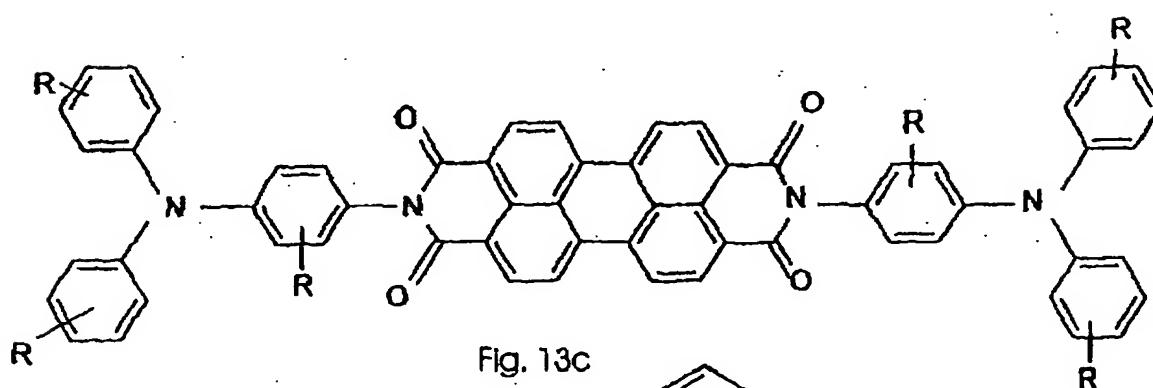


Fig. 13c

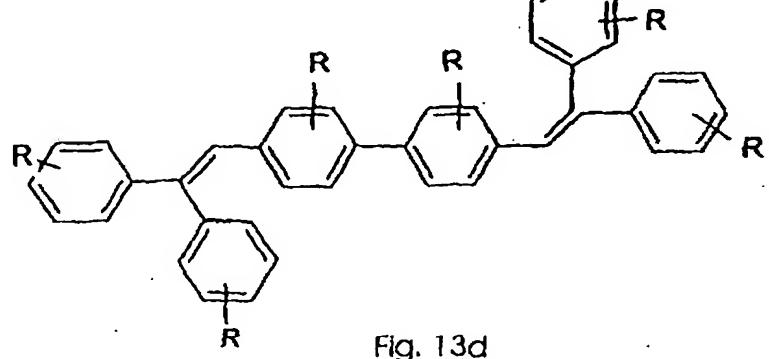


Fig. 13d

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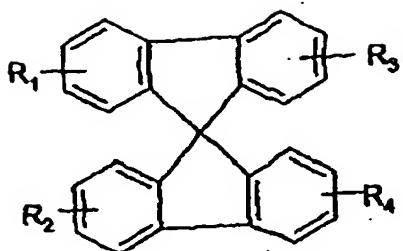


Fig. 14a

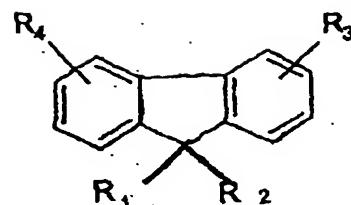
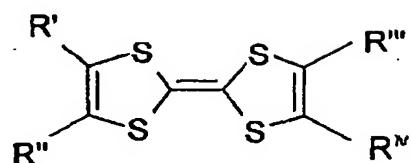


Fig. 14b



OR

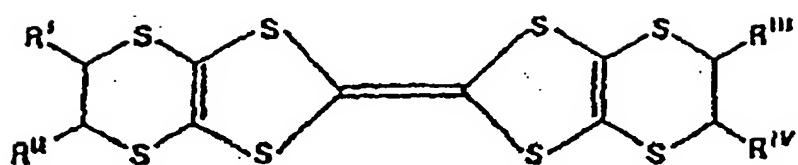


Fig. 14c

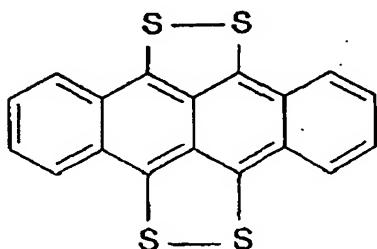


Fig. 14d

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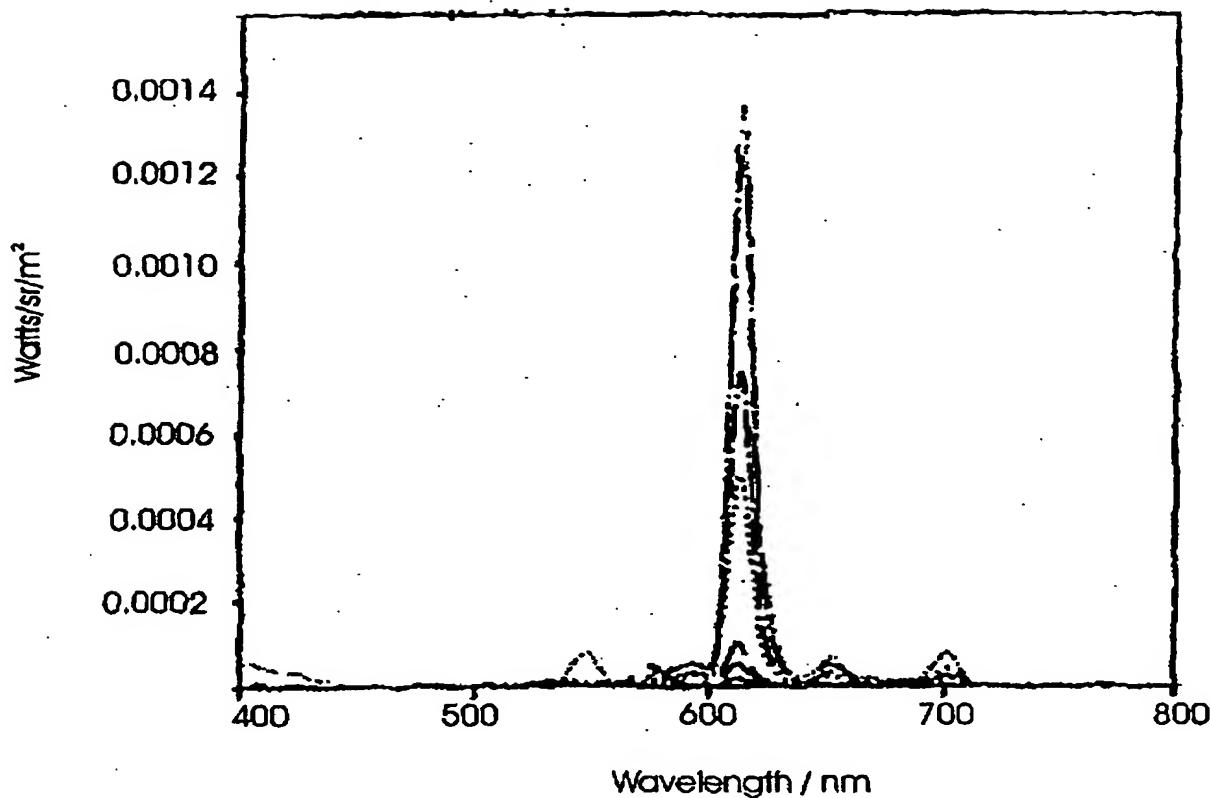


Fig. 14

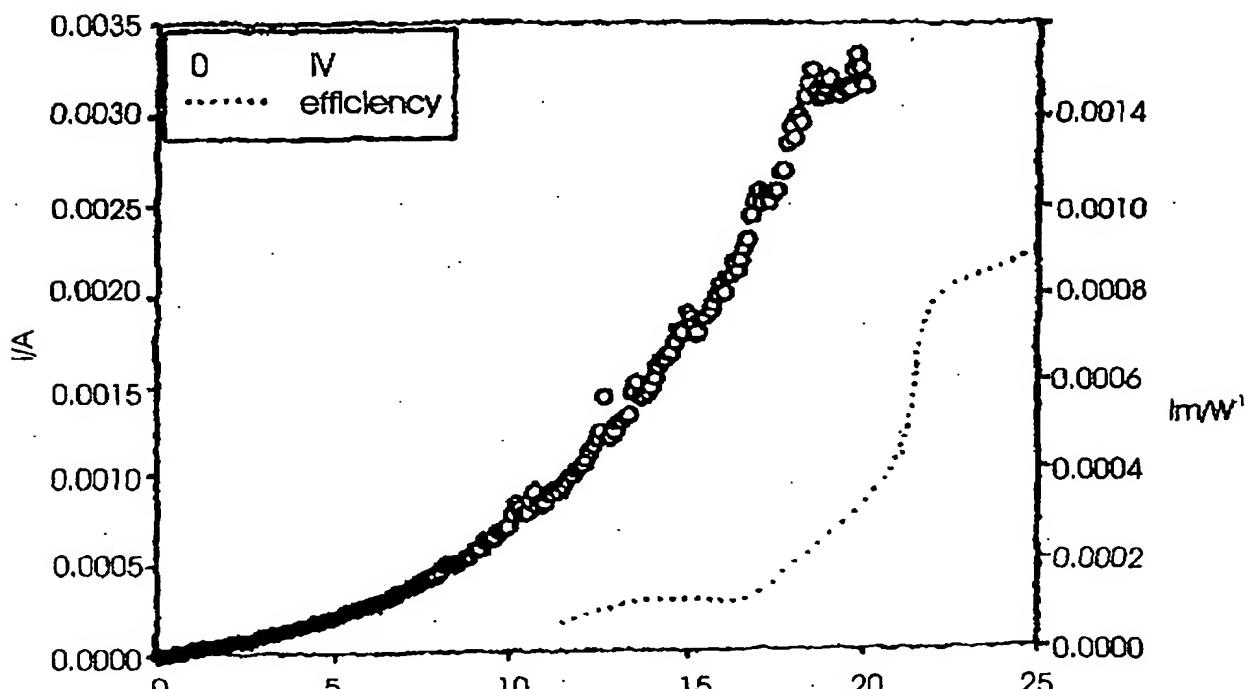


Fig. 15

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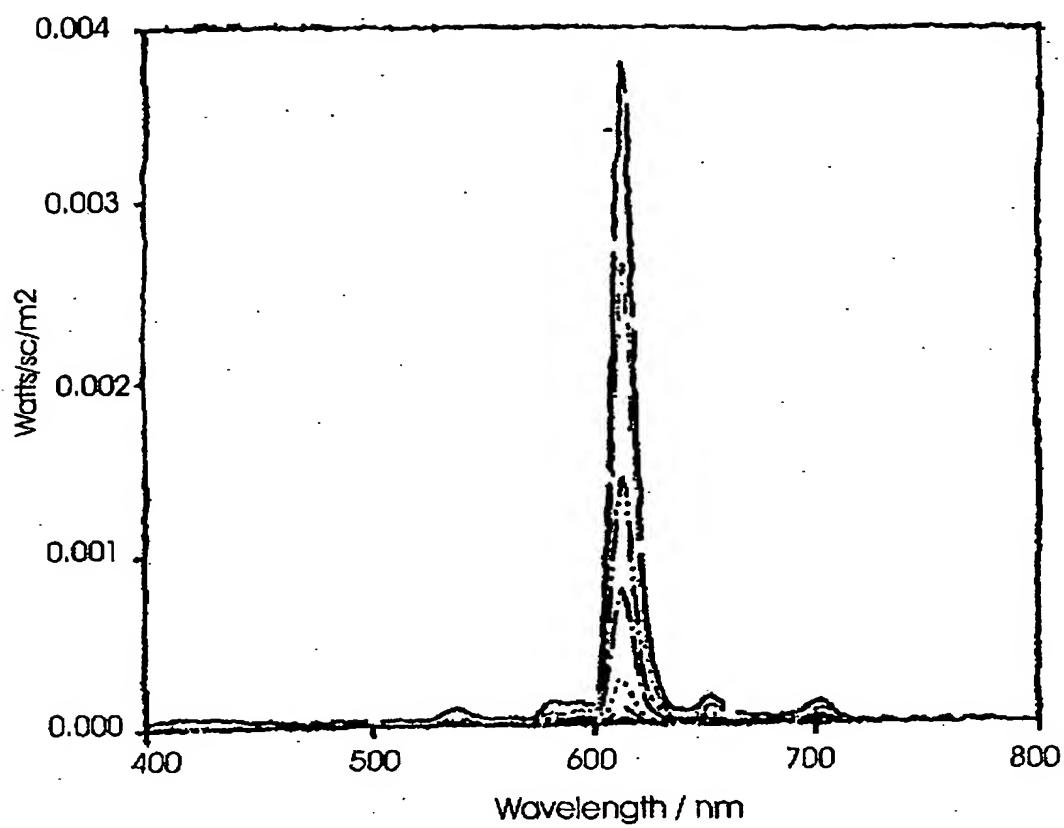


Fig. 16

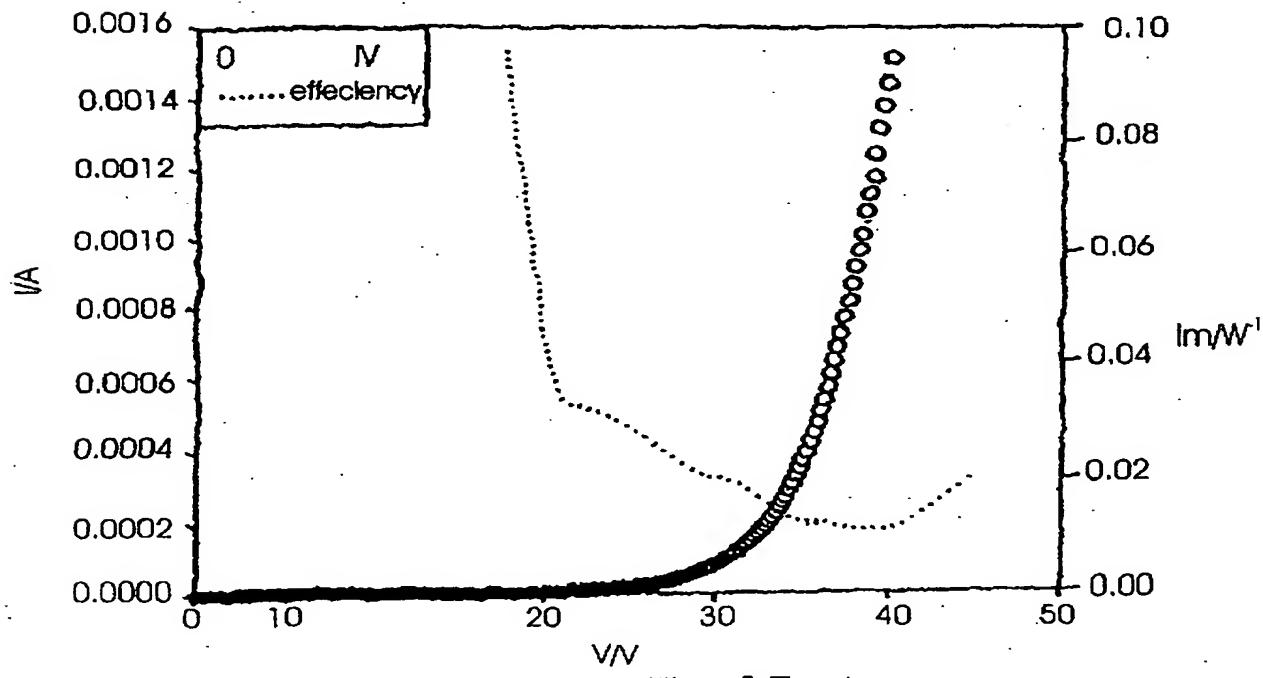


Fig. 17

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ITO / DFDA(5nm) / M-MTDATA(30nm) / Yellow(55nm) / LiF(0.7nm) / Al(150nm)
11-1337

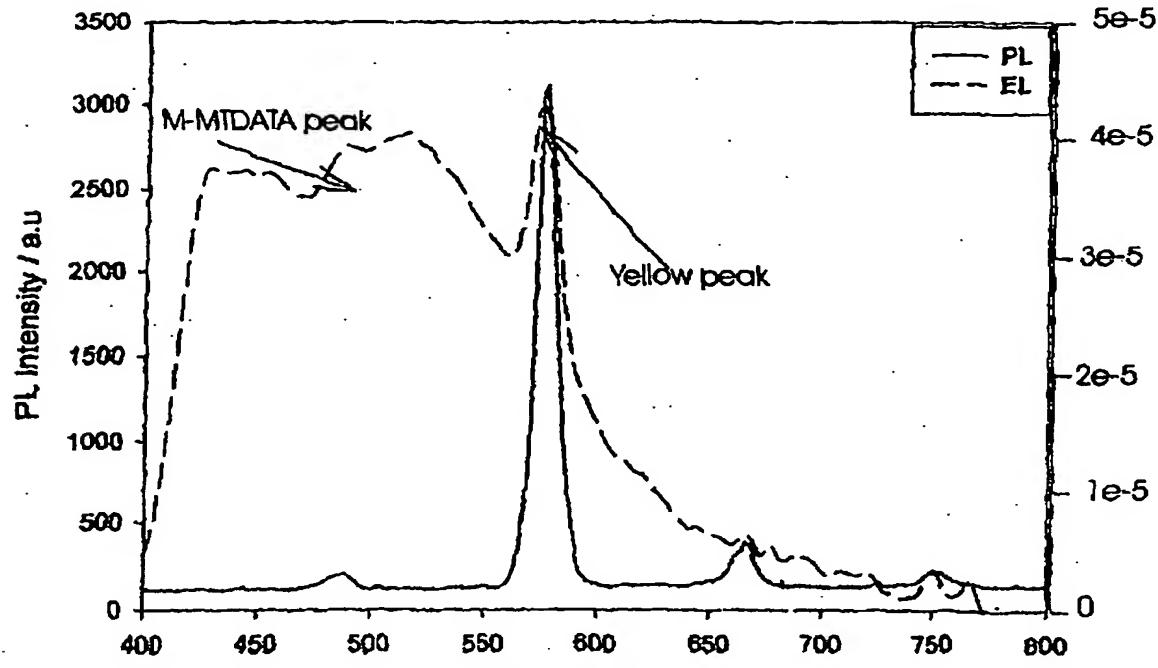


Fig. 18

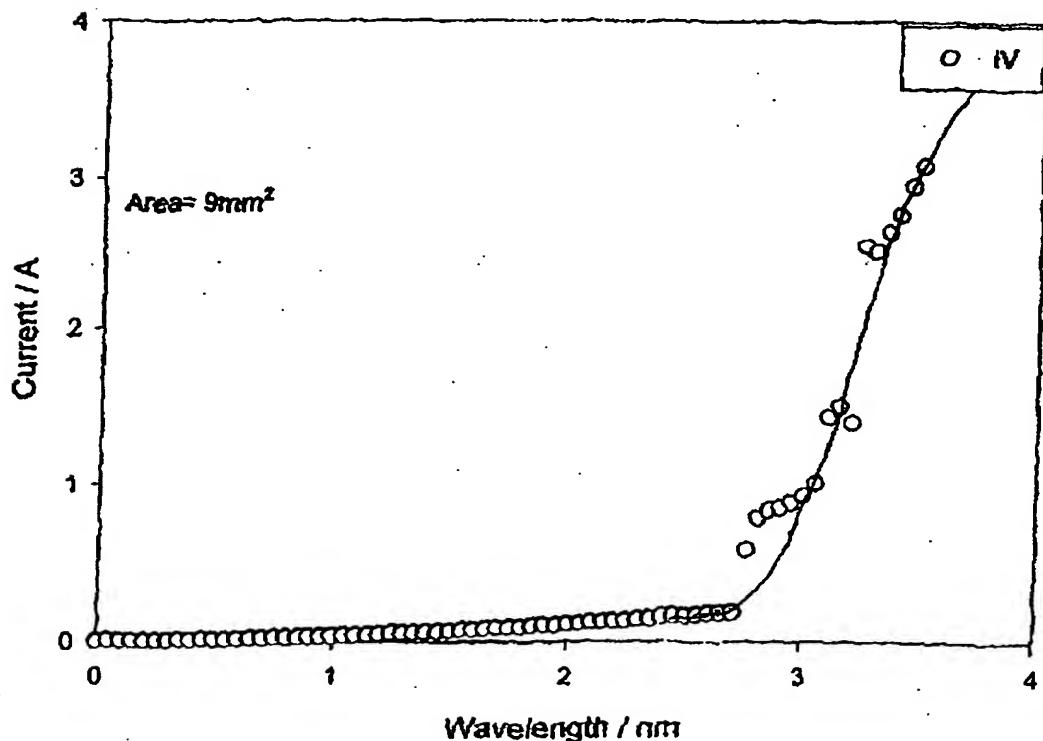


Fig. 19
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INTERNATIONAL SEARCH REPORT

Intern

Application No

01/05111

A. CLASSIFICATION OF SUBJECT MATTER
 IPC 7 H05B33/14 H01L51/20 C09K11/06

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
 IPC 7 H05B H01L C09K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 00 32719 A (KATHIRGAMANATHAN POOPATHY ;SOUTH BANK UNIV ENTPR LTD (GB)) 8 June 2000 (2000-06-08) cited in the application	1-3, 7-10, 12-14, 16,21, 22,25 23
Y	example 3	
X	LIANG C J ET AL: "Organic electroluminescent devices using europium complex as an electron-transport emitting layer" PREPARATION AND CHARACTERIZATION, ELSEVIER SEQUOIA, NL, vol. 359, no. 1, 24 January 2000 (2000-01-24), pages 14-16, XP004321459 ISSN: 0040-6090 the whole document	1-3, 7-10, 12-14, 16,18,25
		-/-

 Further documents are listed in the continuation of box C. Patent family members are listed in annex.

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- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the International filing date
- "L" document which may throw doubt on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the International filing date but later than the priority date claimed

"T" later document published after the International filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"A" document member of the same patent family

Date of the actual completion of the International search

31 January 2002

Date of mailing of the International search report

07/02/2002

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Authorized officer

Lehnert, A

INTERNATIONAL SEARCH REPORT

Intern

Application No

GB 01/05111

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 00 32718 A (KATHIRGAMANATHAN POOPATHY ;SOUTH BANK UNIV ENTPR LTD (GB)) 8 June 2000 (2000-06-08) cited in the application the whole document	1-3, 7-10, 12-14, 16,21, 22,25
X	WO 98 58037 A (KATHIRGAMANATHAN POOPATHY ;SOUTH BANK UNIV ENTPR LTD (GB)) 23 December 1998 (1998-12-23) cited in the application page 16; claims; example 7; table 2	1-3, 7-10,16, 18,21, 22,25
X	WO 00 44851 A (KATHIRGAMANATHAN POOPATHY ;SOUTH BANK UNIV ENTPR LTD (GB)) 3 August 2000 (2000-08-03) cited in the application page 7, line 9 - line 13 page 5, line 7 - line 9; examples 1,2	1,2,7,9, 10,16, 18,25
X	WO 00 26323 A (KATHIRGAMANATHAN POOPATHY ;SOUTH BANK UNIV ENTPR LTD (GB)) 11 May 2000 (2000-05-11) cited in the application the whole document	1,2, 7-10,16, 18,25
Y	WO 00 32717 A (KATHIRGAMANATHAN POOPATHY ;SOUTH BANK UNIV ENTPR LTD (GB)) 8 June 2000 (2000-06-08) cited in the application claim 9; figure 1; example 8	23
X	LING Q ET AL: "A novel high photoluminescence efficiency polymer incorporated with pendant europium complexes" POLYMER, ELSEVIER SCIENCE PUBLISHERS B.V., GB, vol. 42, no. 10, May 2001 (2001-05), pages 4605-4610, XP004232603 ISSN: 0032-3861 the whole document	1-3,7,8, 10
A	WO 99 53724 A (UNIV PRINCETON ;UNIV SOUTHERN CALIFORNIA (US)) 21 October 1999 (1999-10-21) figure 1	1-3, 7-19, 21-23,25
A	US 5 837 391 A (UTSUGI KOJI) 17 November 1998 (1998-11-17) column 1, line 42 - line 44; figure 1	11

FURTHER INFORMATION CONTINUED FROM PCT/SA/ 210

Continuation of Box I.2

Claims Nos.: 4-6, 20, 24

Present claims 4-6 relate to an electroluminescent device comprising an electroluminescent layer comprising a metal complex with at least two metal atoms as part of the complex.

Present claim 20 relates to an electroluminescent device comprising a layer in which the whole transporting material and the light emitting material are mixed.

Present claim 24 relates to an electroluminescent device comprising a layer in which an electron transmitting material and a light emitting metal compound are mixed to form one layer

The claims cover all products having this characteristic, whereas the application provides no support within the meaning of Article 6 PCT and/or disclosure within the meaning of Article 5 PCT.

Consequently, a search has not been carried out for those claims 4-6, 20, 24 which do not appear to be supported and disclosed.

Present claims 1-3, 7-19, 21-23, 25 relate to an extremely large number of possible products. Support within the meaning of Article 6 PCT and/or disclosure within the meaning of Article 5 PCT is to be found; however, for only a very small proportion of the products claimed. In the present case, the claims so lack support, and the application so lacks disclosure, that a meaningful search over the whole of the claimed scope is impossible. Consequently, the search has been carried out for those parts of the claims which appear to be supported and disclosed, namely those parts relating to the products prepared in the examples and closely related homologous compounds.

The applicant's attention is drawn to the fact that claims, or parts of claims, relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure.

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